DEVELOPMENT OF AN IMPROVED DOSE RECONSTRUCTION SYSTEM FOR THE GENERAL POPULATION AFFECTED BY THE OPERATION OF THE MAYAK PRODUCTION ASSOCIATION

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Progress Report on Project 1.1

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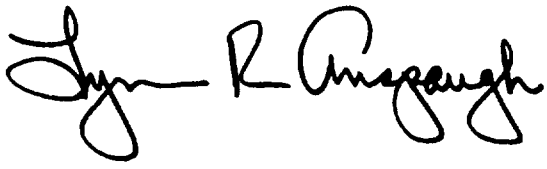

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BACKGROUND

History of the Mayak Complex

The Mayak Production Association (MPA) was the first facility in the former Soviet Union for the production of plutonium for nuclear weapons (Fetisov et al. 1993; Akleyev and Lyubchansky 1994; Fetisov 1996) and is located north of the City of Chelyabinsk. Construction of the MPA facility began in 1945; startup of the plant occurred in June 1948 with the operation of the initial uranium-graphite reactor for the production of plutonium. The first radiochemical plant was placed on-line in December 1948, and the production of enough plutonium for an initial weapons-related test soon followed. There was also a plant for "standard plutonium production" (Koshurnikova et al. 1996). Over the intervening years a total of seven reactors have operated at this site; five uranium-graphite reactors have now been decommissioned. The two remaining reactors are operated mainly for the production of isotopes. Since 1977 the radiochemical plant has been used extensively to reprocess fuels from power reactors and from transport and research reactors. The radioisotope plant dates back to 1962 and is now one of the major suppliers of sources and preparations of radionuclides. Other major activities include the Instrument Engineering Plant, the Repair and Machine Shop, the Central Research Laboratory (CRL), and the Experimental Scientific Research Centre.

Significant worker and population exposure in the nearby Urals region occurred as a result of failures in the technological processes in the first facilities of the MPA in the late 1940's and early 1950's. Members of the public were exposed via major discharges to the Techa River and to the atmosphere. In addition one of the waste-storage tanks exploded in 1957 with another major release. The Mayak CRL undertook early radiation monitoring and determination of exposures. The major work that has been done in reconstructing the identity and quantity of material released to the Techa River was performed by staff of the CRL during the 1950's.

Environmental Releases

The major sources of environmental radioactive contamination were the discharges of about 10^{17} Bq of liquid wastes into the Techa River (1949–1956); an explosion in the radioactive waste-storage facility in 1957 (the so-called Kyshtym Accident) that formed the East Urals Radioactive Trace (EURT) due to dispersion of 7.4×10^{16} Bq into the atmosphere; and gaseous aerosol releases (about 2×10^{16} Bq of ^{131}I in total) within the first decades of the facility's operation.

The majority of the releases occurred in the first years of facility operation, when the waste-management facilities were still being developed. Low- and intermediate-level liquid wastes were routinely released; these routine releases, however, were not the only source of environmental contamination. The Mayak Facility C (radioactive waste-storage facility) was the site of both the Techa River releases and the EURT release. The first available description of the Techa River releases is in the report of the Alexandrov Commission, a special governmental panel created after the discovery of high environmental contamination in 1951. (This report itself is still classified.) In mid-1951, high levels of contamination were found in canals

returning supposedly clean cooling water to the Techa River. Studies showed a correspondence of pulses of large amounts of radioactive materials with application of cooling water to a particular series of tanks. These tanks were operated in a manner somewhat analogous to those at the U.S. Hanford Site: Liquid wastes came from multiple sources within the Mayak processes, wastes cascaded from one tank to another, and complex chemical interactions produced solids that scavenged certain radionuclides in some tanks and freed them in others. Thus, a fraction of the Techa River release was soluble and another fraction was associated with particles. Therefore, the mixture of radionuclides released is very difficult to determine. Soon after the discovery of the uncontrolled and unmonitored releases the low- and intermediate-level discharges, and the other cooling water, were diverted into Lake Karachai.

Exposure of the Population

There were many villages on the Techa River that were downstream from the MPA when the discharges occurred to the river. Villagers were exposed according to a variety of pathways. (A more complete discussion of all pathways that have been considered is detailed in Degteva et al. 1996a.) The more significant pathways included drinking of water from the Techa River, external gamma exposure due to proximity to the Techa River bottom sediments and shoreline, and use of the Techa River water for irrigation of food crops. In addition some members of the Techa River Cohort may have been exposed to the gaseous emissions from the MPA, although this is believed to be a relatively minor source of exposure. After the extent of the major contamination of the Techa River became known, several villages on the upper part of the Techa River were evacuated. Villagers on the lower part of the Techa have remained in their homes up through the present time.

Unique Opportunities

Both the Mayak Worker and the Techa River Cohorts are unique in that members have received unusually high doses, but at low-to-moderate-dose rates. It is likely that study of these populations will provide the best opportunities to determine whether a dose-rate-reduction factor exists for the induction of cancer in human populations. The Techa River Cohort is one of a few that represents an unselected population; the presence of two distinct ethnic groups also provides the opportunity to examine the population variability of risk factors.

The foundation of the dose reconstruction for the Techa River Cohort is also unique. *Over half of the members of the cohort have had whole-body counter measurements of direct relevance to the dose reconstruction.*

Preliminary Studies

Studies of the possible effects of radiation on those exposed to the releases to the Techa River were started in Russia in the 1950s. Russian and United States scientists have been involved in collaborative research programs since 1995.

Epidemiologic studies. Medical checkups of people living in the Techa Riverside communities had been started by 1951. In 1955 a specialized medical institution, known as Specialized Dispensary No. 1, was established in order to determine the health status of the exposed population. This institution became known later as Branch No. 4 of the Institute of Biophysics and is now the Urals Research Center for Radiation Medicine (URCRM). In 1968 the Techa River Registry was created with the goal of including residents of the Techa Riverside villages who lived there during the periods of high exposure from 1949 through 1952. The registry includes data on 26,500 such residents; the registry also contains data on 29,700 persons exposed in utero and/or the progeny of exposed parents and on 7,800 persons who were late entrants exposed after 1952

A preliminary report on the status of the follow-up of the Techa River Cohort has been published by Kossenko et al. (1997). It is reported that, despite a number of limitations, there does appear to be an increasing risk of mortality from leukemia and other cancers with increasing radiation dose.

Dose-reconstruction studies. Systematic measurements of radioactive contamination in and near the Techa River started in the summer of 1951. The contamination of the river water, bottom sediments, flood-plain soils, vegetation, fish, milk, and other food stuffs, and external gamma-exposure rates were measured.

The population of the contaminated territories was chronically exposed to external and internal irradiation. In addition to medical examinations, individual data on the conditions of contact with the contaminated river (the distance of the house from the water's edge, the source of drinking water, fishing, etc.) were collected. Also, radiometric measurements of bioassay and autopsy samples were performed. All places and terms of residence inside the contaminated area were collected for the members of this registry for the purposes of individual-dose reconstruction. Also, extensive measurements of ^{90}Sr content in teeth were performed beginning in 1960 and in forehead bone beginning in 1976; whole-body counting for ^{90}Sr has been performed since 1974; at this time over half of the members of the Techa River Cohort have had at least one whole-body count.

The basis of the past dose-reconstruction efforts for the Techa River Cohort has been summarized in several publications, including Degteva and Kozheurov (1994); Degteva et al. (1994, 1996b, 1997a); Kozheurov (1994); Kozheurov and Degteva (1994). The absorbed doses due to external exposure were estimated on the basis of systematic measurements of gamma-exposure rate along the banks of the river and the typical life-style patterns of the inhabitants of the riverside villages. This approach has given the average annual absorbed doses from external sources for different age groups in each village. A major activity of the current project is to provide this information on an individual basis.

Several efforts have been undertaken in an effort to validate the external doses. One study was undertaken in the now evacuated Village of Metlino. Samples of bricks from abandoned buildings were collected, the quartz was extracted from the bricks, and dose was assessed by using the quartz as a thermoluminescent dosimeter (Bougrov et al. 1995). The

results were doses ranging from 0.76 to 5.28 Gy in the outer cm of the bricks. The highest dose was from a brick wall located near the Techa River and the Metlinsky Pond. Another study (Romanyukha et al. 1996) was performed using electron paramagnetic resonance (EPR) measurements of teeth collected from current or former residents. Teeth were collected from 86 inhabitants of the town of Kamensk-Uralsky, which is not located on the Techa River, in order to determine the age-dependent contribution of background to tooth-enamel dose. EPR measurements of teeth and ^{90}Sr whole body counts were performed on 22 residents of the middle and lower Techa Riverside communities, and absorbed doses were measured in teeth from five residents of the upper Techa.

Results of the above mentioned studies and historical evidence indicates that the main contributor to internal exposure among the radionuclides released into the Techa River was ^{90}Sr, which is accumulated in bone tissues and retained for many years. In vivo beta-ray measurements on teeth, which have been performed since 1960, and a large number of ^{90}Sr measurements in whole body have been the basis of internal dose reconstruction (Kozheurov 1994). The reconstruction of internal dose depends on both estimates of the intake and models for metabolism of ingested radionuclides. Beta-ray measurements on teeth are utilized to deduce the annual levels of intake of ^{90}Sr in the different villages in the different age cohorts. The ingestion of other radionuclides (^{89}Sr and ^{137}Cs predominantly) occurred mostly with water in the first three years of the river contamination. The intake rates of ^{89}Sr and ^{137}Cs were therefore derived from estimates of the ingestion of ^{90}Sr scaled in terms of the radionuclide composition of the river water. These data were used to estimate age-dependent intake rates for all Techa River villages (Kozheurov and Degteva 1994). Calculation of absorbed doses in tissues due to radionuclide incorporation is based on age-dependent metabolic and dosimetric models and the corresponding ingestion rates. A large number of measurements of ^{90}Sr-body content made with a whole-body counter (WBC) has been utilized for the validation of the metabolic model for strontium retention in human bone (Degteva and Kozheurov 1994). Absorbed doses in red bone marrow (RBM) and bone surfaces (BS) have been calculated for all age cohorts. The absorbed doses in RBM and BS are substantially higher than those in other tissues, because ^{90}Sr was the main radionuclide of interest and strontium is a bone-seeking element. The upper limit of total doses absorbed in RBM is estimated as about 3 Gy.

THE CURRENT STUDY

This project is a comprehensive program to develop improvements in the existing dosimetry system for the members of the Techa River Cohort by providing more in-depth analysis of existing data, further search of existing records for useful data, model development and testing, evaluation of uncertainties, verification of procedures, and validation studies of current and planned results. This current project is the result of the first year's pilot study (Degteva et al. 1996a) and extensive meetings and discussion among the participants in the dosimetric and epidemiologic studies. The details of the project have been specified in the proposal document (Degteva et al. 1996c). Following approval of the proposed work it was necessary to provide a revised list of Tasks and Milestones. This latter list was reproduced as Appendix 1 in the March 1998 Progress Report (Degteva et al. 1998a).

The specific aim of this project is to enhance reconstruction of external and internal radiation doses for approximately 26,500 individuals in the Techa River Cohort. The purpose of the enhanced dose reconstruction is to support companion epidemiologic studies of radiogenic leukemia and solid cancers (NCI-RERF-URCRM Project and JCCRER Project 1.2). The current database of preliminary individual doses will be expanded and upgraded, and the uncertainty in the doses reconstructed will be evaluated.

The most recent joint meeting of the Russian and American team members took place in April 1998 in Chelyabinsk. About half of the time was spent in preparing for and participating in a review meeting for the Russian and U.S. Scientific Review Groups. The remainder of the time was spent in completing preparation of publications for submission to peer-reviewed journals, review of progress on various tasks, and initiation of the task on analyzing the uncertainty in the doses now being calculated and the doses that will be calculated in the future. In addition, Drs. Degteva and Anspaugh had an opportunity to meet and discuss the project at a meeting sponsored by the European Commission and the U.S. National Cancer Institute in Germany during November 11–13, 1998.

The next joint meeting of the Russian and American team members will take place in April 1999* in Chelyabinsk and in Moscow (the Moscow meeting is an international scientific meeting sponsored by the JCCRER and the European Commission; April 28–30, 1999). While some of the time at Chelyabinsk will be spent in preparation for the Moscow meeting, most of the time will be devoted to continuing work on the Project. During the remainder of Fiscal Year 1999 it is anticipated that Drs. Degteva and Kozheurov will make a joint trip to the U.S. to continue project work and that Drs. Anspaugh and Napier will make a trip to Russia (if it is more appropriate and timely, this trip may take place in early Fiscal Year 2000).

Progress on Task 1:

Task 1. Feasibility analysis of the development of a special system for obtaining tooth samples from the Techa River residents (M. Degteva).

Subtask 1. Examine the feasibility of establishing a system of tooth collection for the members of the Techa River Cohort. This system should be oriented toward the collection of teeth as they are being extracted for dental health purposes. A special aim would be the collection of teeth from former residents in the Upper Techa River locations that have long since been evacuated.

This work was completed with delivery of the Milestone 5 Final Report (Degteva et al. 1997b), which was included as Appendix 3 in the March 1998 Progress Report (Degteva et al. 1998a).

* The long time between formal meetings of the project participants was not planned; it is due to unanticipated budgetary events during Fiscal Year 1998.

Progress on Task 2:

Task 2. Source-term and environmental data analysis (M. Vorobiova).

Subtask 2. Check the consistency of available source-term and historical monitoring data in order to verify and confirm limited available information on the source term.

Subtask 3. Develop a simple empirical river model in order to verify distance dependence of radionuclide composition of river water and to link radionuclide concentrations in water and bottom sediments with γ-exposure rates on the banks near the river.

This work was completed with the delivery of the Milestone 1 Final Report (Vorobiova et al. 1997), which was an Attachment to the March 1998 Progress Report (Degteva et al. 1998a). In addition, two manuscripts based upon this Milestone Report have been prepared and submitted to *Health Physics* (Vorobiova et al. 1999; Vorobiova and Degteva 1998). These two manuscripts were reproduced as Appendices 1 and 2 of the October 1998 Progress Report (Degteva et al. 1998b). Since then the report on historical monitoring of Techa River contamination (Vorobiova et al. 1999) has been accepted and the galley proofs, which are reproduced here in Appendix 1, have been returned. The second report (Vorobiova and Degteva 1998) has been reviewed, and the modified manuscript has been returned to *Health Physics.*

Progress on Task 3:

Task 3. External dose reconstruction (D. Burmistrov)

Subtask 4. Verify γ-exposure rates on the banks near the river and validate accumulated doses calculated on the basis of verified γ-exposure rates in specific points of the upper and middle Techa by the results of thermoluminescent measurements in bricks from the same sites.

Subtask 5. Enter into computer and analyze available data on the outdoor and indoor γ-exposure rates on the territories of the Techa Riverside settlements in order to determine the dependence of γ-exposure rate from the distance from the edge of the water and to verify the contributions to the total external dose from the sources of radiation exposure in the streets, gardens, and houses.

Subtask 6. Evaluate organ-specific absorbed doses from external exposure (based on literature data) and include these values into the TRDS system code.

Subtask 7. Validate absorbed doses in individuals from external sources of radiation by the results of electron paramagnetic resonance measurements in teeth.

Routine work on this task is continuing. Some results were included in the Milestone 1 Report (Vorobiova et al. 1997); additional results are reported in the Milestone 4 Report (Bougrov et al. 1999), which is an attachment to this report. With the publication of the Milestone 4 report, Subtask 4 above is completed. Dr. Burmistrov is currently on a Fellowship Program at Harvard; this will delay the completion of the remainder of this task by a few months.

Progress on Task 4:

Task 4. Internal dose reconstruction (V. Kozheurov and E. Tolstykh).

Subtask 8. Verify radionuclide-ingestion levels derived from the results of river modeling (Subtask 3) and include into TRDS Module 1 the ingestion levels of other radionuclides from the releases (Zr, Nb, Ru, Ce, etc.).

Subtask 9. Include into TRDS Module 2 appropriate metabolic and dosimetric model calculations (based on literature data) for other radionuclides according to the results of Subtask 8.

Work on this task was completed with the submission of the Milestone 3 Final Report (Tolstykh et al. 1998), which was included as an Attachment to the October 1998 Progress Report (Degteva et al. 1998b).

Progress on Task 5:

Task 5. Uncertainty analysis (M. Degteva, L. Anspaugh, and B. Napier)

Subtask 10. Enter into computer "interview data" (source of drinking water, the distance of the house from the edge of water etc.) for the residents of two Techa Riverside villages: Metlino and Muslyumovo.

Subtask 11. Develop an algorithm of classification (grouping) of the persons belonging to "families" (households) according to the source of drinking water and/or the distance from the edge of water based on individual-strontium measurements, family-member lists (available in data base MAN) and interview data (Subtask 10).

Subtask 12. Perform the grouping of persons according to the algorithm from Subtask 11 for the residents of Metlino and Muslyumovo villages. Evaluate "inside group" and "between groups" contributions into variation for internal dose (based on whole-body counter measurements). Evaluate the feasibility of estimating the contributions to external dose uncertainties from the distance of the house from the river and individual variability in behavioral regimes

Subtask 13. Evaluate the contributions to internal dose uncertainties from the source of drinking water (river or wells) and individual variability in diet habits and metabolic parameters on the basis of Metlino and Muslyumovo data.

Subtask 14. Evaluate the feasibility of the reconstruction of household specific doses for the entire Techa River Cohort on the basis of the results of Subtasks 12 and 13. Develop a plan for further study on dose reconstruction.

Accurate quantification of the dose-response function for populations exposed to ionizing radiation requires appropriate treatment of the uncertainty in the dosimetry and the uncertainty in

measurements of the response. Uncertainty in the determination of either the dose or biological effect (cancer mortality, for example) obviously can influence the derivation of the dose-response function for an exposed cohort. Until recently, uncertainty in dosimetry has been given limited attention in risk analysis. A dose estimate for any specified individual may be affected by two types of errors: random and systematic. Random errors are those that vary independently among exposed individuals. Systematic errors are those that cause an uncertain bias to an entire subgroup of an exposed cohort. The examination of sources and magnitudes of uncertainty in radiation doses to individuals in an exposed cohort is an important task of any dosimetric study.

According to Task 5 we plan to investigate the influence of such factors of systematic errors as "the source of drinking water (river or wells) in the period of massive releases (1950–1951)." Data from interviews of the Techa Riverside residents about the sources of drinking water are used for this purpose. Such interviews were carried out in the 1950s by members of special teams from the Moscow Biophysics Institute and in the 1970s by staff of the URCRM. The results were recorded in out-patient cards kept in the URCRM archives. The first step of work (Subtask 10) was the entry into a computer database of the "interview data" for two major villages on the Techa Riverside, Metlino and Muslyumovo, in order to evaluate the completeness and quality of these data for the upper and middle Techa region. This data base was described in our March 1998 Progress Report (Degteva et al. 1998a). During this year information on the large Techa Riverside settlement, Brodokalmak (located in 109 km from the site of release), was added to the data base in order to obtain a larger set of data for statistical analysis.

Data base description. Computer files were prepared for each of the three settlements that have the following fields for each person: Identification code, year of the first interview, and source of drinking water in 1950–1951 (river, well or both). Also, these files contain the code of the household in which each person lived in 1950–1951 and the address inside the settlement (street, house number), if such data are available in copies of old taxation books. Table 1 illustrates the data from these files.

Table 1. Data on water supply and joint living in one and the same house in the period of major radioactive releases 1950–1951.

	Metlino	Muslyumovo	Brodokalmak
Number of Techa River Cohort members permanently residing in 1949–1951	1221	2481	3354
Number of out-patient cards available	1196	2426	3319
Number of persons examined who responded about the source of drinking water	530	712	1004
Number of persons with information in copies of taxation books	817	2014	2436
Number of persons with available address (street, house number)	713	1253	2421

As seen from Table 1 out-patient cards were available for 99% of the total number of persons who lived in the investigated villages in 1950–1951. Among them 56% of the Metlino residents (upper Techa) and only 30% of Muslyumovo and Brodokalmak residents (middle Techa) were questioned about the source of water supply (it should be noted that some people questioned in the 1970s did not remember their source of drinking water in 1950–1951). Information on other persons in the household is available for 81% of the Muslyumovo residents, 73% of the Brodokalmak residents and 67% of the Metlino residents. Address data that allows the determination of the position of the house relative to the river are available for 72% of the Brodokalmak residents, 58% of the Metlino residents and, 51% of the Muslyumovo residents.

Assessment of data on water supply. According to historical data, there were 13 wells in Metlino Village for 1221 residents, only four wells in Muslyumovo for 2481 residents, and 28 wells in Brodokalmak for 3354 residents.

For Metlino 439 persons (83% of the total number of questioned residents) were examined in 1951–1955. The examinations of people in the middle Techa region were started in 1953, and 418 persons from Muslyumovo (59%) and 558 persons from Brodokalmak (56%) were investigated in 1953–1963. These "early data" seem to be the more reliable, because the people seem to remember reliably their source(s) of drinking water. The next peak of information was collected during the period 1974–1980, when the Techa Riverside residents filled out special questionnaires at the same time that they were being measured with the URCRM whole body counter (WBC). Additional data from 76 Metlino residents (14%), 277 Muslyumovo residents (39%) and 398 Brodokalmak residents (40%) were obtained in this period. These "late data" are less reliable, because people tended to forget what sources of drinking water they used 26–30 years ago.

The results of analysis of the questionnaire data, summarized in Table 2, show that the maximal percent of "drinkers" (users of the river water) is observed for the Muslyumovo settlement and the minimal percent for the Brodokalmak settlement. This appears to be in agreement with historical data on the number of wells in these two settlements.

In order to assess the reliability of the "interview data," they were matched with the WBC results as adjusted for the age and time dependencies. It was found that WBC data are available for 42% of Metlino residents, 53% of Muslyumovo residents, and 45% of Brodokalmak residents (who responded about their water supply). The distributions of WBC values for the "drinkers"

Table 2. Data from interviews of the Techa Riverside residents about the sources of drinking water in 1950–1951.

Source of water supply	Number of persons		
	Metlino	Muslyumovo	Brodokalmak
River	360 (68%)	553 (78%)	397 (40%)
Wells	138 (26%)	128 (18%)	457 (46%)
River + wells	32 (6%)	31 (4%)	150 (12%)

(persons who used river water) and "non-drinkers" (persons who used well water) are shown in Figs. 1-3, and some statistical characteristics are presented in Table 3.

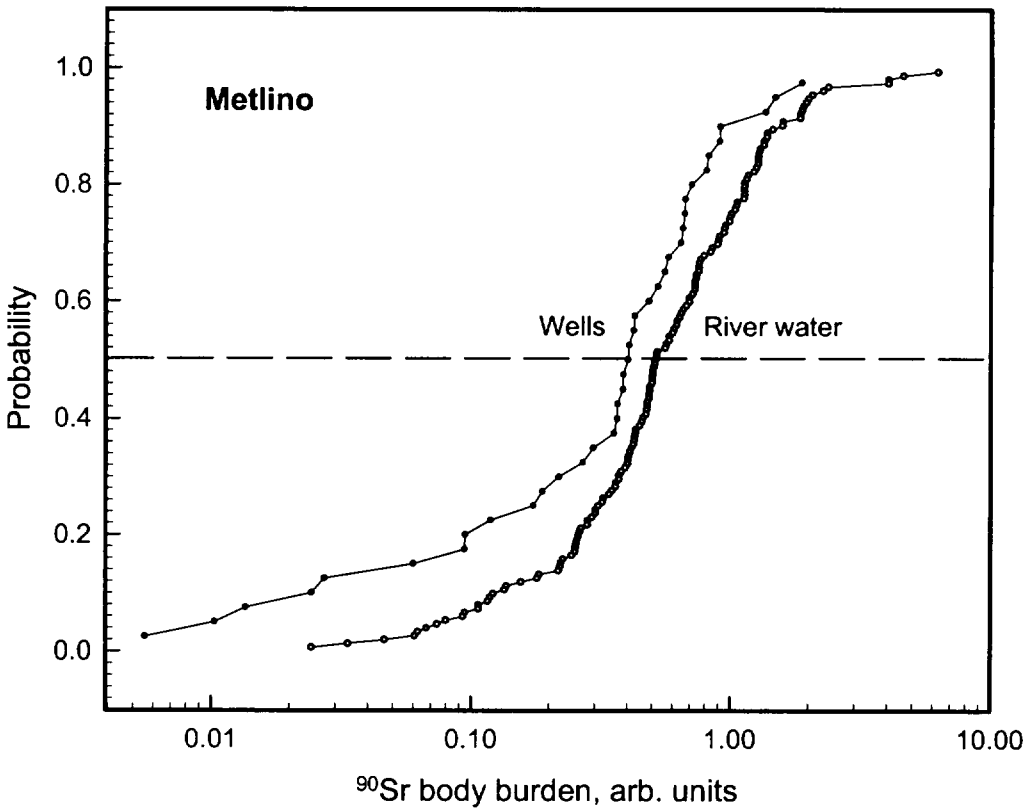

Fig. 1. Distributions of ^{90}Sr-body burdens in "drinkers" and "non-drinkers" of river water for residents of Metlino.

Table 3. Statistical characteristics of the distributions of relative WBC values (corrected for the age and the time of measurements) as functions of the place of residence and the source of drinking water.

Settlement:	Metlino		Muslyumovo		Brodokalmak	
Source of water:	River	Wells	River	Wells	River	Wells
Number of persons measured	153	49	291	72	135	245
Minimum	0	0	0	0	0	0
Maximum	6.25	1.88	5.05	4.28	2.67	1.34
Median	0.51	0.37	0.98	0.74	0.26	0.04
Mean	0.77	0.41	1.08	0.92	0.38	0.15
Standard error	0.07	0.06	0.04	0.10	0.04	0.02

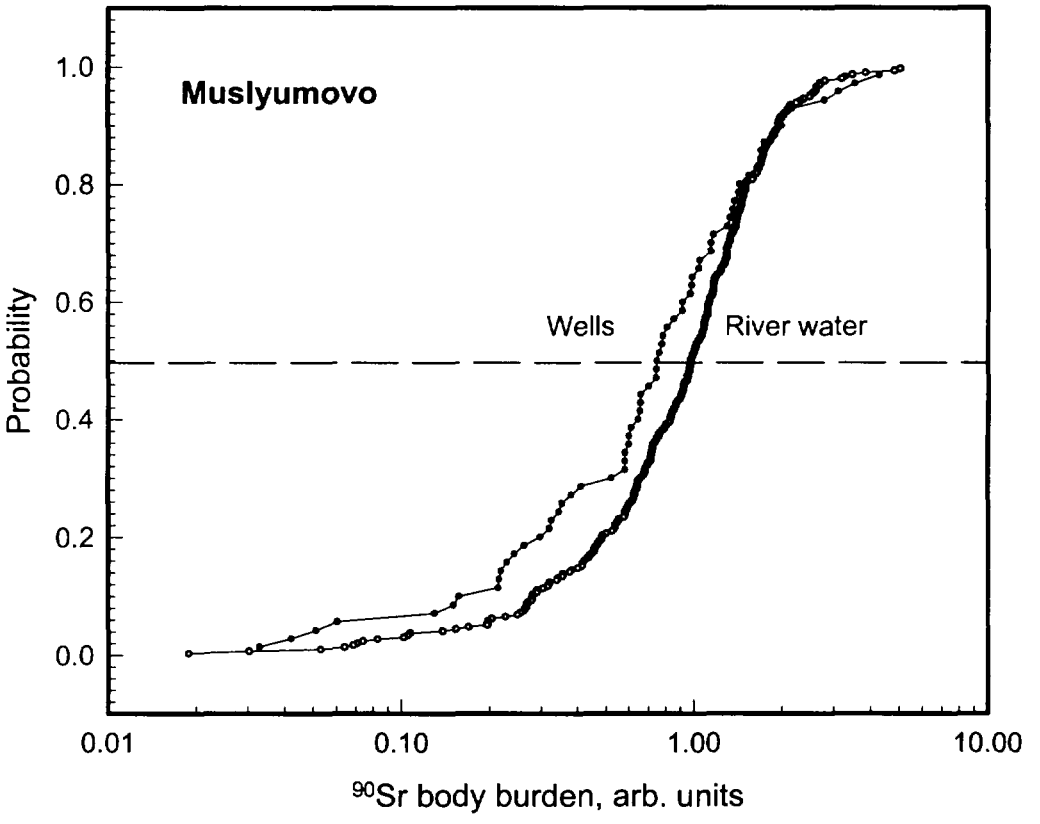

Fig. 2. Distributions of ^{90}Sr-body burdens in "drinkers" and "non-drinkers" of river water for residents of Muslyumovo.

Statistical analysis of these data shows that there is a significant difference between groups of "drinkers" and "non-drinkers" for Metlino and Brodokalmak. As for Muslyumovo, the difference between the two groups is not statistically significant; this implies that almost all residents of this settlement have permanently or episodically used river water for drinking during the period of massive radioactive releases.

In spite of the existence of the difference between groups, individual classification of persons on the basis of "interview data" will have great uncertainties because the ranges of individual-body burdens for "drinkers" and "non-drinkers" have significant overlap. This could be explained by two factors:

- Significant variability of individual metabolic parameters; and
- The existence of some mistakes in the data of interview. The roots of such mistakes could be psychological or methodological, but apparently the percent of such mistakes was similar for all samples of persons examined, as the differences between statistical characteristics of groups of "drinkers" and "non-drinkers" are logical.

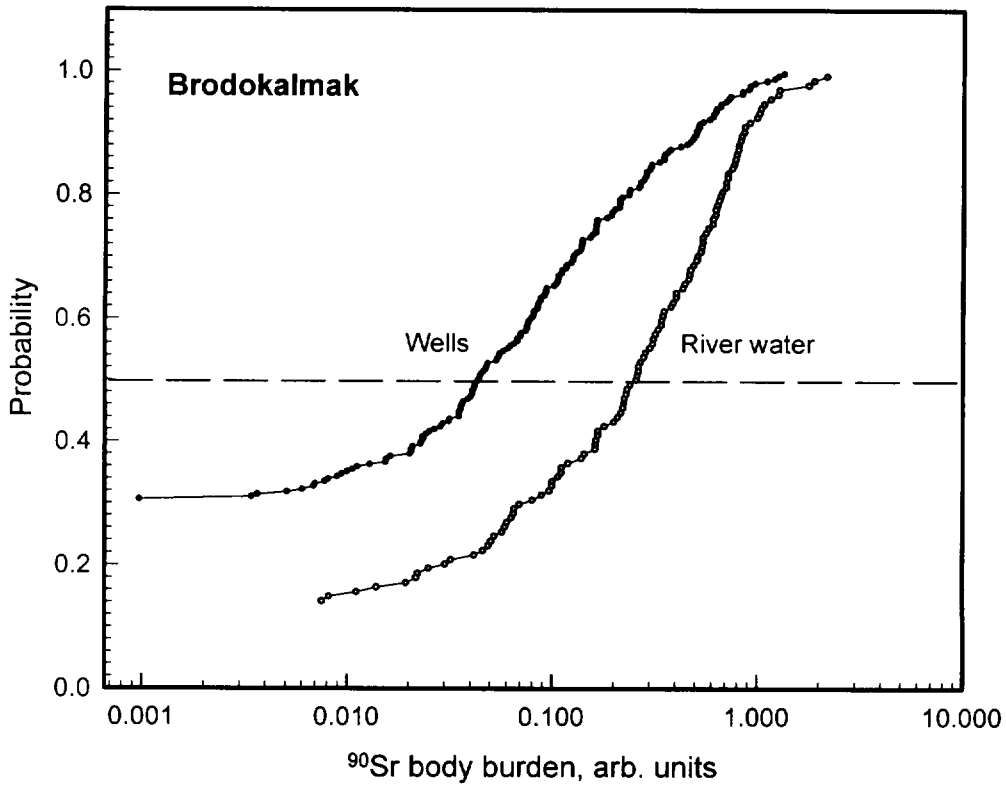

Fig. 3. Distributions of ^{90}Sr-body burdens in "drinkers" and "non-drinkers" of river water for residents of Brodokalmak.

Preliminary analysis of uncertainty for the TRDS model. The method being used in the upgrade to the Techa River Dosimetry System (TRDS-98) basic dose calculation is relatively simple; it can be written in a single equation:

$$C_0 = \sum_{y=1}^{Death} \sum_{L} \frac{M_{yL}}{12} \left[\left(\sum_{r} 365 * I_{YrL} * DF_{ro} \right) + \frac{A_o}{24} \left(D_{Riv,L,Y} * T_1 + D_{Out,L,Y} * T_2 + D_{In,L,Y} * (24 - T_1 - T_2) \right) \right]$$

where

C_o = Cumulative dose to organ o (Sv);
Y = Year (1949–1960);
L = River-location identifier;
M_{YL} = Months in year Y spent in location L;
12 = Months per y;
r = Radionuclide identifier;
354 = Days per y;
$I_{Y,r,L}$ = Intake function for year Y, radionuclide r, and location L (Bq d^{-1});

$DF_{r,o}$ = Dose-conversion factor for radionuclide r in organ o (function of age, related to Y (Sv Bq^{-1});
A_o = Air-to-organ correction, independent of energy (Sv Gy^{-1});
24 = Hours per day;
$D_{Riv,L,y}$ = Air dose rate by river at location L in year Y (Gy y^{-1});
$D_{Out,L,Y}$ = Air dose rate in streets at location L in year Y (Gy y^{-1});
$D_{In,L,Y}$ = Air dose rate indoors at location L in year Y (Gy y^{-1});
T_1 = Time spent on riverbank (h d^{-1}); and
T_2 = Time spent outdoors (h d^{-1}).

In this formulation, the term $M_{Y,L}$ comes from individual-life history information and is a series of constants; all the other terms are either calculated or approximated, and have some uncertainty associated with them.

The source of information for each term in the TRDS was evaluated. The terms T_1 and T_2, while ideally coming from individual surveys, are currently assigned default values, depending on whether the individual is a child (<7 years old) or adult in year Y. The dose factors DF_{ro} are derived from the URCRM metabolic model for ^{90}Sr and are taken from ICRP publications for other radionuclides. The external dose rates $D_{Riv,L,y}$, $D_{Out,L,y}$, and $D_{In,L,y}$ are derived from the radionuclide contents of sediment calculated from the model of Vorobiova and Degteva (1999) and the radiation-transport model of Lappa and Burmistrov (1994); or alternatively, from tabulations of standard external dose-rate factors, such as those provided in Eckerman and Ryman (1993). The key term $I_{Y,r,L}$ is derived from information in Kozheurov and Degteva (1994). It is the product of the appropriate factors in table 1, table 3, and table 4 of that report. As such, it has a very complex uncertainty structure, particularly since table 1 is further based on the doctoral thesis of Skryabin (1971).

In order to estimate the uncertainty of the dose estimates calculated using the TRDS, a stochastic Excel™ spreadsheet was developed using the Crystal Ball™ Monte Carlo utility. The spreadsheet implements the TRDS equation for the years 1950 through 1955, for external doses and internal dose from ^{90}Sr only, for five Techa River villages. For this implementation of the spreadsheet, individuals are assumed to be born in 1950. The spreadsheet provides results for the annual and 5-year cumulative doses for individuals born in the year 1950 in villages of Metlino, Nadirov Most, Muslyumovo, Brodokalmak, and Pershino. These are respectively 7, 48, 78, 109, and 212 km from the Mayak release point. The doses at Metlino are dominated by the external component, those at Muslyumovo and further downstream are primarily internal, and at Nadirov Most are a combination.

The doses estimated using the spreadsheet are reasonably close to those estimated with the full TRDS system for the very limited number that it handles. The radiation doses at Metlino have a large external component, giving them a generally normal uncertainty distribution with a standard deviation of 30–50%. The doses at Muslyumovo and villages further downstream are smaller, but dominated by the internal component, and have a more pronounced lognormal distribution, with a geometric standard deviation of about two.

For the external doses, the parameters for time on the waterfront (T_1), exposure-to-dose ratio (A_o), and dose rate (D_{riv}) appear to have the larger influences on the estimates of dose. These are most important at Metlino, because of the high external dose rate, and are less important downstream. For the internal doses, the primary source of uncertainty in the spreadsheet as currently set up is the dose-conversion factor normalization, for both effective and red bone marrow dose. In part, this is because a single normalization factor (geometric standard deviation of 2) is used for all ages. However, this is a reasonable approximation, because the intake-to-dose factor should be correlated for any given individual as a function of age. This result is comparable with results of other analyses [e.g., Farris et al. (1994)]; the variability between individual responses to a given intake is frequently as large as the variability in the predicted environmental concentrations or intakes. The next most important parameter for internal dose is the intake function I_{YrL}. Within this function, the food-intake rate as a function of age [table 1 of Kozheurov and Degteva (1994)] is important, which as noted above is complex and based on the thesis of Skryabin. Recent work by Burmistrov[†] has indicated that the Kozheurov and Degteva table 3 component of the intake function may have a higher uncertainty than initially considered as well. This is an area of future refinement for this work, which is now in its preliminary phase.

Progress on Task 6:

Task 6. Whole-body-counter calibration and modification (V. Kozheurov and A. Kovtun).

Subtask 15. Manufacture an anthropomorphic physical phantom of the body of an adult with an uniform distribution of ^{90}Sr in the skeleton. This phantom will be used for the recalibration of whole-body counter SICH-9.1. The design of this phantom was described in the Final Report of JCCRER Project 1.1 (Degteva et al. 1996a).

Subtask 16. Prepare the protocol and perform the calibration of whole-body counter SICH-9.1 for ^{90}Sr, ^{137}Cs and ^{40}K using special phantoms. Evaluate instrumental errors for SICH-9.1 in order to provide better verification of the thousands of whole body counts available at the URCRM.

Subtask 17. Develop a mathematical phantom to simulate the spectral and angular distribution of photon radiation, including bremsstrahlung, at the surface of the phantom resulting from the radioactive decay of incorporated radionuclides in order to study the effects of non-uniform distribution of ^{90}Sr in the different bones and structures of the skeleton and to study the effects of variations in individual-body geometry.

Subtask 18. Develop the design for modification of SICH-9.1 and perform the upgrade of SICH-9.1 in order to replace obsolete detectors and electronics and provide continuity of whole body measuring.

[†] Personal communication, 1999.

Subtasks 15 and 16. Work on these two subtasks has been completed and is documented in the Milestone 2 Final Report (Kozheurov et al. 1998), which was included as an Attachment to the October 1998 Progress Report (Degteva et al. 1998b).

Subtask 17. Now that subtasks 15 and 16 have been finished, more attention will be devoted to Subtask 17. It is our intent to transfer an existing mathematical phantom from the U.S. to the URCRM. The phantom for this purpose has been selected, but it does not include bremsstrahlung as a source emission. Modification to the computer phantom will be necessary, but has not been completed. Transfer of the phantom to Dr. Kozheurov is planned for a visit of Drs. Degteva and Kozheurov to the U.S. within the next several months.

Subtask 18. Work on upgrading WBC SICH-9.1 consists of two parts:

- To specify and purchase a new set of detectors and electronic system for spectrum analysis; and

- To replace computer controlling scanning bed system (mechanical and electronic parts).

The University of Utah sent out a request for bid package in early 1998. One bid was received from "Pribory Oy," an official distributor of EG&G ORTECH. However, this bid included a Value Added Tax of 20%, which put the bid over the amount of money available for the purchase. As agreements between the governments of the U.S. and the Russian Federation were interpreted as exempting the payment of such tax, requests were made to EG&G ORTECH to secure a letter of exemption from this tax. This letter of exemption was not received, and the existing bid expired.

The University of Utah reissued the Request for Bid. The winning bid was received from EG&G ORTECH, and the order was placed in November 1998. We have been informed that most of the equipment is now on hand in Moscow; after the detectors themselves are received it is anticipated all equipment will be shipped to Chelyabinsk and installed at the URCRM within the next few months.

Other work to upgrade the WBC, called "computer controlled scanning bed system," was reported on in the March 1998 Progress Report (Degteva et al. 1998a). This work has been completed.

Progress on Task 7:

Task 7. Electron paramagnetic resonance (EPR) measurements (A. Romanyukha).

Subtask 19. Perform about 90 measurements of teeth for the residents of the middle Techa in order to evaluate age dependence of EPR signal for exposed people and compare individual external doses based on these measurements with the results of calculations (Subtask 7).

Subtask 20. Perform about 90 measurements of teeth for the residents of non-contaminated areas of the Ural Region in order to evaluate age dependence and the nature of background EPR signal.

Progress on this Task was reported on extensively in the March 1998 Progress Report (Degteva et al. 1998a) and at the Scientific Review Groups' meeting in April 1998 in Chelyabinsk. In July 1998 work on EPR dosimetry at the University of Utah was cancelled by the DOE, and support was also withdrawn for work to be done in Russia.

The Final Report (Haskell 1998) on EPR dosimetry that included work performed by the Russian investigator was included as an Attachment to the October 1998 Progress Report (Degteva et al. 1998b).

Progress on Task 8:

Task 8. Luminescence measurements (N. Bougrov).

Subtask 21. Perform TL measurements for about 7–10 sites in Muslyumovo Settlement in order to evaluate the distribution of radiation fields and to compare external doses based on these measurements with the results of calculations (Subtask 4).

Progress on this Task was reported on extensively in the March 1998 Progress Report (Degteva et al. 1998a). In July 1998 work on thermoluminescence dosimetry at the University of Utah was cancelled by the DOE, and support was also withdrawn for work to be done in Russia.

The status of work that had been performed on this Task was summarized in Appendix 4 of the October 1998 Progress Report (Degteva et al. 1998b). The material in the mentioned Appendix 4 was a preprint of a paper submitted to *Health Physics;* this paper has now been published (Bougrov et al. 1998) and a reprint is included in Appendix 2.

The Final Milestone Report No. 4 (Bougrov et al. 1999), which includes the work on Task 8 and Subtask 4 from above, has been completed and is included as an Attachment to this Progress Report.

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APPENDIX 1

Galley Proofs

Vorobiova, M. I.; Degteva, M. O.; Burmistrov, D. S.; Safronova, N. G.; Kozheurov, V. P.; Anspaugh, L. R.; Napier, B. A. "Review of historical monitoring data on Techa River contamination." *Health Phys.* (in press, 1999).

Paper

REVIEW OF HISTORICAL MONITORING DATA ON TECHA RIVER CONTAMINATION

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Abstract—**The Mayak Production Association was the first Russian site for the production and separation of plutonium. The extensive increase in plutonium production during 1948–1955, as well as the absence of reliable waste-management technology, resulted in significant releases of liquid radioactive effluent into the rather small Techa River. This resulted in chronic external and internal exposure of about 30,000 residents of riverside communities; these residents form the cohort of an epidemiologic investigation. Analysis of the available historical monitoring data indicates that the following reliable data sets can be used for reconstruction of doses received during the early periods of operation of the Mayak Production Association: Temporal pattern of specific beta activity of river water for several sites in the upper Techa region since July 1951; average annual values of specific beta activity of river water and bottom sediments as a function of downstream distance for the whole river since 1951; external gamma-exposure rates near the shoreline as a function of downstream distance for the whole Techa River since 1952; and external gamma-exposure rate as a function of distance from the shoreline for several sites in the upper and middle Techa since 1951.**

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Key words: contamination, environmental; plutonium; exposure, population; monitoring, environmental

INTRODUCTION

The Mayak Production Association (MPA) was the first Russian site for the production and separation of plutonium. This plant began operation in 1948, and in its early days technological failures resulted in the release of large amounts of radioactive effluent into the rather small Techa River. The residents of the riverside communities were exposed to chronic external and internal irradiation. Extensive monitoring efforts for the environment and the population at this site began in 1951. The "Techa River Cohort" (TRC) has been studied for several decades by scientists from the Urals Research Center for Radiation Medicine (URCRM). An increase in both leukemia and solid tumors with radiation dose has been noted (Kossenko et al. 1997). This finding suggests that, with continuing improvements in the quality of the follow-up and dosimetry, study of the TRC has the potential to provide quantitative estimates of the risks of stochastic health effects produced by chronic low-dose-rate radiation exposure for the general population. These population-based risk estimates would have relevance to the regulation of radiation exposure throughout the world.

The current dose-reconstruction system (known as the Techa River Dosimetry System or TRDS) for the TRC, which numbers about 30,000 people, is grounded firmly on whole-body counts for half of the members of the cohort (for the evaluation of internal dose, which was mainly due to incorporated ^{90}Sr) and on direct measurements of external gamma-exposure rates (Kozheurov 1994; Degteva et al. 1998). A joint Russian-American project has now been implemented under the auspices of the U.S.–Russian Joint Coordinating Committee on Radiation Effects Research (JCCRER). This project is concerned with a comprehensive program to develop improvements in the existing dosimetry system for the TRC members by providing more in-depth analysis of existing data, further search of existing records for useful data, model development and testing, evaluation of uncertainties, verification of procedures, and validation studies of current and planned results.

A set of conceptual models that defines the relationships, pathways, and parameters that form the basis of the dose-reconstruction efforts has been described (Degteva et al. 1996a). The hierarchy of information required for calculating doses to people who lived along the Techa River also has been described by Degteva et al. (1996a). One of the important tasks of the project is the analysis of available historical data on source terms of the releases and on environmental contamination.

The purposes of this paper are to

- Describe historical data regarding the operating history of the MPA and available measurements of releases;

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- Present accumulated historical Techa River data on hydrology, sediment loading, and dam-construction history; and
- Compile and evaluate available data on radionuclide concentrations in river water and sediments and gamma-exposure rates on the banks.

THE MAYAK COMPLEX AS A SOURCE OF TECHA RIVER CONTAMINATION

Construction of the MPA was begun in 1948 in the Southern Urals for the purpose of producing plutonium for nuclear weapons (Fetisov 1996). This complex consisted of a series of uranium-graphite reactors operating with thermal neutrons and using direct flow water-cooling loops (Complex A); a radiochemical plant for the extraction of ^{239}Pu from uranium irradiated in the reactors (Complex B); a chemical-metallurgical plant for metallic plutonium production and machining (Complex V); and radioactive waste-management and storage facilities (Complex C). Over the intervening years six reactors operated at the MPA for the production of weapons-grade plutonium. Of these, five were graphite moderated while the sixth was originally heavy-water moderated (Novosselov and Tolstikov 1995). The graphite-moderated reactors have now been shut down (Fetisov 1996); the heavy-water moderated reactor was later modified to become a light-water moderated reactor, and it remains in operation today. A seventh reactor is operational for the production of isotopes for civilian uses. The operating histories of the plutonium-production reactors from 1949 through 1967 are shown in the Appendix. Since 1977, the radiochemical plant has been used extensively to reprocess fuels from power, transport, and research reactors.

The extensive increase in plutonium production during 1948–1955, as well as the absence of reliable waste-management technology, resulted in significant releases of liquid radioactive effluent into the Techa River. The chronology of major events connected with the evolution of the exposure situation on the Techa River is shown in the Appendix. These data were extracted from several sources (Marey et al. 1953, 1954, 1956, 1965; Ilyin 1956; Alekseeva et al. 1957; Borovinskikh et al. 1958; Anikin et al. 1959; Marey 1959; Makhonko 1994; Fetisov 1996; Novosselov and Tolstikov 1997).

In 1949, high-level wastes were routed to the tank farms in Complex C, and low-level wastes only were released to the Techa River (Fig. 1). However, in 1950, F1
in order to reduce the volume of material going to the tanks in Complex C, a process for "decontamination" of high-level wastes was introduced, with a portion of the radioactivity directed to the tanks and a portion released to the river. In July 1951, it was discovered that this process did not work as intended, and that during this period high concentrations of radionuclides had been released into the river. Also during this time cooling water from the Complex C tanks was discharged into the Techa River at the same location as the technological

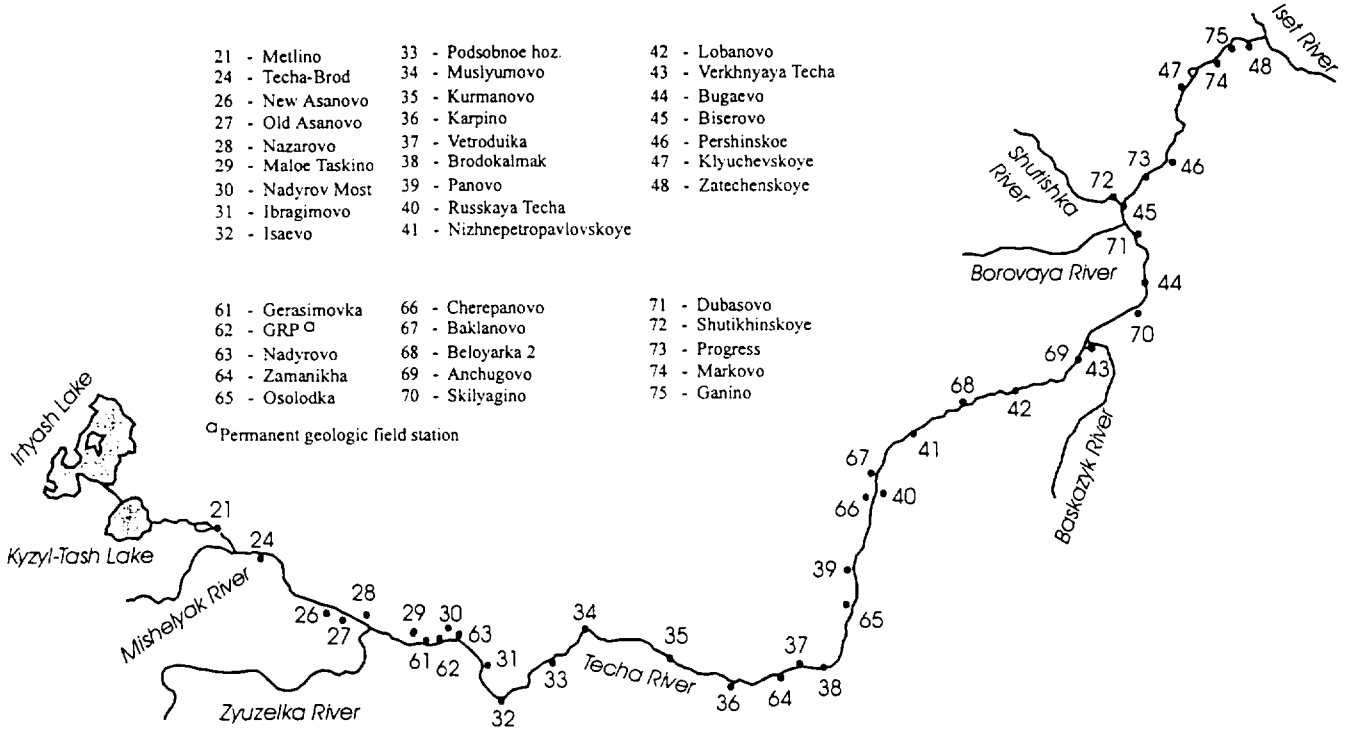

Fig. 1. Schematic map of the Techa River (approximate scale) and of the villages located on its banks before contamination occurred. Villages No. 21–48 served as the sites of historical routine sampling.

wastes. Leaks in the tank-cooling lines caused some of these discharges to be highly contaminated. These "wild releases" were unmonitored and unnoticed until 1951. Over this period, about half of the total release to the Techa River resulted from the technological releases and about half from the wild releases.

In late 1951, several activities to control the releases and to remediate the environmental contamination were begun. The main technological releases were diverted into Karachay Lake (Fig. 2). Large amounts of water were released into the Techa River from Lake Kyzyl-Tash in an effort to wash the contamination out of the area of Metlinsky Pond and reduce the exposure rates. Over the next several years, a series of dams were built in the upper Techa with bypass canals to halt the spread of contamination (Fig. 2a, b, c, and d). The construction of Reservoir 11 and the creation of the bypass canals provided isolation of the most contaminated part of the river bed from the lower parts of the Techa River where the population continued to live. Between 1952 and 1956, residents of all villages within 75 km of the site of the release were resettled in uncontaminated areas. Some of the remaining villages along the Techa River were also resettled between 1956 and 1960. About 7,500 people in all were resettled.

It must be noted that in addition to radiochemical wastes that were the major source of the Techa River contamination, radionuclides entered the river with the water of Kyzyl-Tash Lake used for reactor cooling. Also some surface contamination of the upper Techa region occurred as a result of the Kyshtym accident in 1957, and wind transfer of contaminated silt of Karachay Lake in 1967 (Fig. 2c and d). And, finally, in the period of January–April 1958, Berdyanish Lake waters, which were contaminated as a result of the plume from the Kyshtym accident, were discharged to the Techa River

Fig. 2. Schematic maps of the upper reaches of the Techa River (approximate scale): a) before August 1951; b) 1951–1956; c) 1956–1964; and d) after 1964. Heavy bars indicate dams; hatched lines indicate canals; thin arrows indicate liquid releases; heavy arrows indicate atmospheric releases; the darker areas in c and d indicate the region of ^{90}Sr-deposition density greater than 740 MBq m^{-2} (20 Ci km^{-2}).

(Fig. 2c). Since 1967, the Techa River system has been in a natural self-cleaning regime.

HYDROLOGICAL CHARACTERISTICS OF THE TECHA RIVER

The Techa River (the right tributary of the River Iset) belongs to the basin of the Kara Sea. On the basis of the hydrological characteristics of the river (240 km in length, up to 2 m in depth, mean annual flow rate in the outfall of about 7 $m^3 s^{-1}$), it can be assigned to the category of small rivers. Its main tributaries are the Mishelyak, Zyuzelka, Baskazyk, Borovaya, and Shutishka Rivers (Fig. 1).

The Techa riverside area can be divided into two parts depending on its flood land and bed characteristics:

- In the upper reaches of the Techa, a cascade of hydraulic-engineering constructions are located (Fig. 2), including Reservoirs No. 3 (Koksharov Pond was created at the location of an old weir in August 1951), No. 4 (Metlinsky Pond, which already existed in 1949), No. 10 (created in 1956) and No. 11 (created in 1964). The stretches of the river from Reservoir No. 11 up to the village of Muslymuovo are for the most part swampy, with a poorly marked winding bed overgrown with water plants. The width of the river bed is from 3 to 15 m, and its depth is from 0.5 to 2 m; bed deposits consist of turf-silt or clay;
- In its middle and lower reaches (downstream of the village of Muslyumovo), the river has a well-marked bed, its bottom consisting of layers of sand and slime, and, in some places clay, sand, and gravel. The mean width and depth of the river during the summer time are 22 and 0.5–1 m, respectively.

As the Techa belongs to the type of plain rivers, it has few turnings: The meandering length of the river divided by the length of the straight line from its beginning to its end is about 1.07. The longitudinal profile of the river bed is characterized by the slope of the average line of the bottom and the slope of the water surface along the river course; the midstream and downstream reaches of the river have a slope virtually equal to that of the water surface, i.e., about 0.6 m km^{-1}.

The Techa River receives its supply of water from melting snow and intensive spring floods (Fig. 3). The main source of water supply to the Techa during the summer months is groundwater discharge from water-bearing horizons formed by atmospheric precipitation (Koloskov 1968). During the period of floods, a back-water phenomenon develops due to the river-water effluent that minimizes the amount of groundwater entering the river. On the average, the groundwater penetrating into the river makes up about 10% of the overall river runoff.

A comparison between the mean annual flow rate in the river and the annual level of precipitation in the area

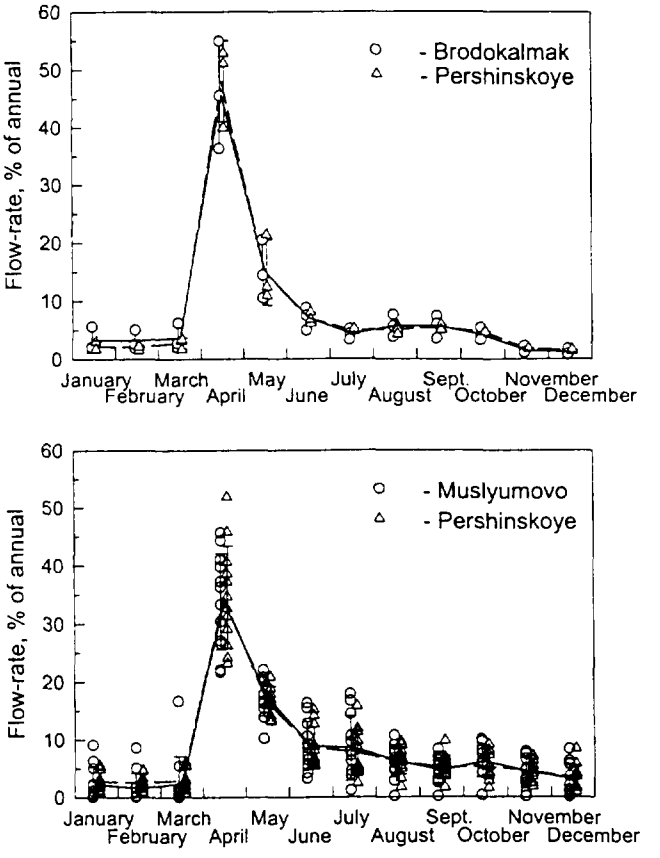

Fig. 3. The Techa River flow rates measured in the following ranges: Brodokalmak-Pershinskoye in 1951–1953 and Muslyumovo-Pershinskoye in 1963–1976. Data were extracted from Shakhov (1967); Starostina (1973, 1974, 1975, 1976); Agapitova (1975); and Katomina (1977, 1979).

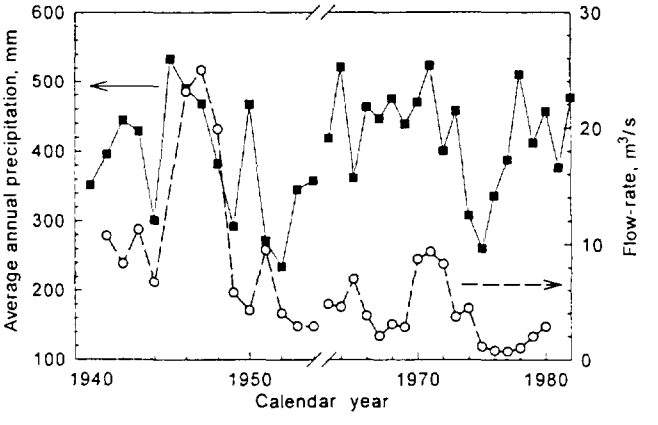

Fig. 4. Mean annual water discharge in the Techa River and annual level of precipitation according to data of Shakhov (1967); Starostina (1973, 1974, 1975, 1976); Agapitova (1975); and Katomina (1977, 1979).

under observation (Fig. 4) has shown that the curve representing the flow rate is actually similar in shape to that referring to precipitation, with a year's delay in dynamics. The installation of the upstream reservoirs in

the mid-1950's changed the flow patterns markedly, as seen in Fig. 4. The flow rate has ranged from 2 to 10 m^3 s^{-1} in recent years. The minimum estimates of flow rate (<1 m^3 s^{-1}) were noted in the drought-afflicted years 1975–1976.

In terms of chemical composition, the water of the Techa River is classified among the carbonate-sodium type; pH = 7.5–8.5; mineralization is estimated to be about 700 mg L^{-1} at present. Ion concentration in the river water does not change significantly with time and averages about 60 mg L^{-1} for Ca^{++}, 30 mg L^{-1} for Mg^{++}, 300 mg L^{-1} for HCO^{-}, and 40 mg L^{-1} for Cl^{-}. The most clear-cut changes after the installation of the last dam (No. 11) were observed in the river-water mineralization level and the content of sulfates in it (Komissarova 1985).

The bed deposits in the upper reaches of the river between Reservoir No. 11 and Muslyumovo consist of layers of turf, silt, and clay. There are flood swamps measuring 300 m to 2 km in width along the river shoreline; the most swampy areas are located between the villages of Nadyrov Most and Muslyumovo at the site where the Zyuzelka River enters the Techa. The central portion of the flood soils is composed of turf-bog soils that give way to meadow-turf ones along the boundaries of the swamps. The thickness of the turf layer ranges from 10 cm to 3 m, and the turf contains a considerable amount of mineral inclusions and increased percentage of ash (10–35%, up to 60% in the near-to-bottom layers). Clay and sandy loam, and less frequently sand, compose the underlying layer of turf.

Bed deposits in the middle and lower reaches of the river are of sandy-silt and sandy-gravel type; the dry flood land measures 200–500 m in width and is composed of turfy-meadow soils. Studies of the mechanical composition of the soils have shown that the sandstone bed deposits and sandy loam soils of the flood meadows are characterized by a higher content of large (0.25–1 mm) particles while the fine fraction (<0.01 mm) is uniformly distributed over the vertical profile with its content determined by the type of soils (Safronova et al. 1986).

AVAILABLE INFORMATION ON DISCHARGE OF RADIOACTIVE MATERIALS INTO THE TECHA RIVER

Information on radioactive releases into the Techa River was analyzed and presented in URCRM Technical Reports (Kozheurov 1985; Shvedov et al. 1990). The primary data on the releases for these reports were extracted from MPA Technical Reports, 1951–53, and the doctoral thesis of D. I. Ilyin (1956) (these documents are kept in MPA archives and are still classified). The extracted information contains data on release dynamics and isotopic composition of wastes. The other sources of information were Technical Reports of the Institute of Biophysics (Marey et al. 1953, 1954, 1956, 1965; Alekseeva et al. 1957; Borovinskikh et al. 1958; Anikin et al. 1959), and the doctoral thesis of A. Marey (1959).

Radioactive effluents were released into the Techa River beginning in March 1949. Initially, high-level wastes were concentrated and kept in specially equipped containers (the tanks of Complex C) and only low-level wastes (after passing through special absorbers) were released into the river. In January 1950, a special facility for the decontamination of high-level liquid radioactive waste was put into operation, and the construction of additional Complex C tanks was stopped. Later it was found that this technology of radioactive waste management was poor and could not give the necessary level of decontamination. As a result of this inadequacy, release rates into the Techa sharply increased. And, in addition to pre-planned releases, some unexpected releases (so-called "wild overflows") were discharged episodically into the river with the cooling waters of Complex C tanks, and the average daily release between March 1950 and November 1951 reached 1.6×10^{14} Bq (4,300 Ci). Starting 28 October 1951, the release of all technological wastes from process operations was routed to Karachay Lake, and only low-level wastes (from laundry, laboratory, and septic systems) continued to enter the Techa River.

All information on the releases may be divided into three parts:

- Mean activity and isotopic composition of releases during specific time intervals (main part). The data contain average values of alpha-, beta- and gamma-emitting activities released and mean percentage of some radionuclides for the time periods March–December 1949, January–February 1950, March 1950–October 1951, and November 1951–December 1952. It is believed that these data were reconstructed by D. Ilyin (the Head of the Mayak Central Laboratory) based on knowledge of technological processes and some measurements that were made in 1951–56. The methods of reconstruction were not described in the available documents;
- Yearly mean activity and isotopic composition of releases in 1953–1956. The data contain average values of beta activities released and mean percentage of main radionuclides and/or groups of radionuclides in 1953–1956. These data were estimated by A. Marey on the basis of Moscow Institute of Biophysics research expeditions (1952–1959); and
- Primary data on activity released in various time periods from 1951 to 1956 (including the information about "wild overflows"). This information contains results of measurements of daily volume and specific beta activity of releases and/or daily activity released, radionuclide composition and percentage of activities on the solids in some days (periods) of the release time. The character of primary data on the release dynamics is presented in Table 1.

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Table 1. Characteristics of primary data on release dynamics.

Period	Values	Character of data	Sources of release
25 September–7 October 1951	Volume and specific beta and gamma activity released	Daily measurements	Determined partially
1952	Volume and/or specific alpha, beta and/or gamma activity released	Episodic measurements	Not determined
March–June 1953	Total beta and/or gamma activity released	Weekly mean	Determined
July–September 1953	Total beta and/or gamma activity released	Monthly mean	Determined

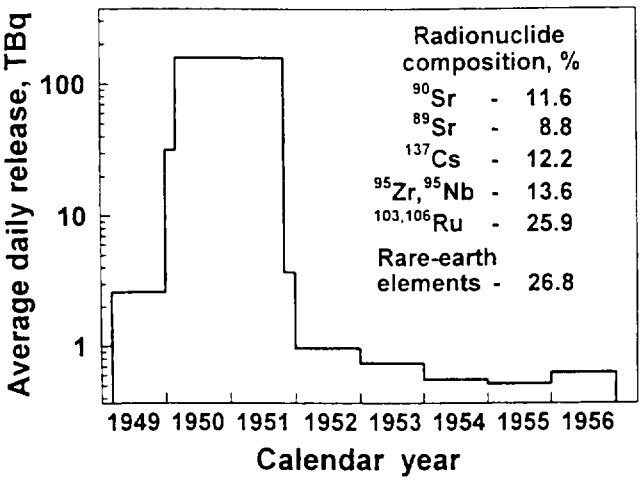

Fig. 5. Average amount of radioactivity released per day into the Techa River between 1949 and 1956 and radionuclide composition (according to the data of the Mayak Central Laboratory, Project Director Dmitry Ilyin) for the time period of major releases (March 1950–October 1951). Data on radionuclide composition at other times are provided in Table 2.

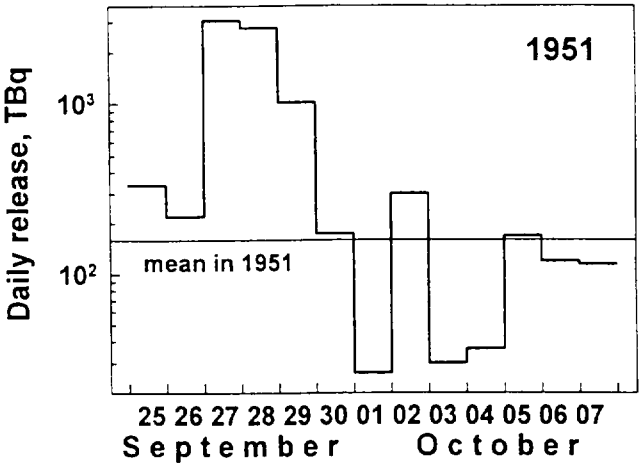

Fig. 6. Fluctuations in release rates observed in autumn 1951.

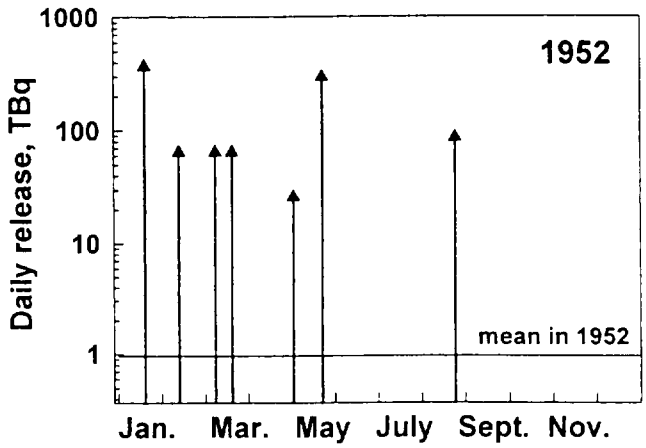

Fig. 7. Peaks of radionuclide releases observed in 1952 relative to annual mean values.

Average release rates evaluated by the Mayak experts under supervision of D. Ilyin are presented in Fig. 5. The total amount of fission products released into the Techa River was about 10^{17} Bq of beta emitters (about 98% of this value was released during 1950–1951). The ratio of gamma-equivalent to beta-activity was about 0.24 gram-equivalent of radium[§] Ci^{-1} (a reporting scheme in use at the time relative to a radium standard). This corresponds to an average holdup time (the time of cooling of irradiated uranium blocks) of about 150 d. The total amount of alpha emitters discharged to the river with wastes was evaluated as less than 1.85×10^{12} Bq (50 Ci). Available data of daily measurements permitted evaluation of the fluctuation of releases in the initial period of monitoring (Fig. 6) and also showed irregularities in rate after the main technological releases were routed to Karachay Lake (Fig. 7).

It was found in 1951 that the releases consisted of suspensions (pH = 7–8) with 3–5 g L^{-1} of sodium nitrate and sodium acetate. Suspended particles consisted mainly of iron hydroxide and organic matter. On average, about 70% of beta activity entered the Techa River with the suspended particles and the remaining 30% was in soluble form.

Radionuclide composition of the releases from the radiochemical plant is presented in Table 2. The values before 1952 should be considered as evaluated theoretically because the first measurements of total activity were started in July 1951, and radiochemical techniques for some radionuclides were not developed until later (in 1953 for Zr + Nb and in 1956 for Ru).

[§] "Gram-equivalent of radium" is an old method used to specify the amount of gamma-emitting materials present in a sample. Thus, one "gram equivalent of radium" is that amount of gamma-emitting radionuclide (or mixture of radionuclides) that produces the same amount of ionization as does one gram of radium in equilibrium with its short-lived progeny. One gram-equivalent of radium is approximately equal to 0.5 μGy m^2 s^{-1}.

Table 2. Radionuclide composition of the releases in different years according to different sources. Due to process changes the release rates of radioactive materials decreased substantially in later years. The *relative* amounts released of radiostrontiums increased due to the long half life of ^{90}Sr and the high solubility of strontium.[a]

Time period	Radionuclide composition, %							Reference
	$^{89}Sr + {}^{90}Sr$	^{89}Sr	^{90}Sr	^{137}Cs	REE	$^{103,106}Ru$	$^{95}Zr + {}^{95}Nb$	
1949		1.8	4.1	11		55.6	30	Ilyin (1956)
January–February 1950		6.9	15.3	21.2	5.7	45.3	9	Ilyin (1956)
March 1950–October 1951		8.8	11.6	12.2	26.8	25.9	13.6	Ilyin (1956)
November 1951–December 1953	25.6–58.0			4.5–15	10–61		8–25	Ilyin (1956)
1954	44.0			10.0			1.0	Marey (1959)
1955	39.0			12.0				Marey et al. (1956)
1956	53.5			23.8	12.5	7.3		Marey et al. (1957)

[a] Notes: 1) All data presented correspond exactly to the original references;
2) REE—rare-earth elements;
3) Radionuclide composition for the period 1949–51—assessed data;
4) For the period November 1951–December 1953—minimal-maximal percentage;
5) For 1954–1956—data of radiochemical analyses.

Center between two columns. See example, attached.

Table 3. Radionuclide distribution in liquid waste sampled on 25 September 1951, before and after centrifugation.

Type of sample	Beta activity, kBq L^{-1}	Concentration of radionuclides, %				
		$^{89,90}Sr$	^{137}Cs	REE	$^{103,106}Ru$	$^{95}Zr + {}^{95}Nb$
Original suspension	44.4	20.2	2.8	47.4	7.6	22.0
Solution (supernatant)	13.7	49.2	9.0	3.5	8.3	30.0
Sediment	30.7	7.3	0	67.0	7.3	18.4

Note: REE—rare-earth elements.

There was only one experimental analysis of the radionuclide composition of the early wastes (on 24–25 September 1951), and this should not be interpreted as representative for the whole period of acute exposure. The results of this analysis, presented in Table 3, show that almost all cesium, 75% of strontium, and about 50% of zirconium and niobium were in soluble form, but about 98% of rare earth elements entered the river absorbed on suspended particles. The measured radionuclide composition is different from "theoretical" values. The ratio of gamma-equivalent to beta-activity for the composition calculated using a holdup time of 150 d gives the value of about 0.15 g-equivalent of radium Ci^{-1}. The measurements of beta- and gamma-activity for the release on 25 September 1951 gave a ratio equal to 0.48; that is three times more than the calculated value. Such a discrepancy suggests the existence of some systematic errors in the technique for measuring gamma activity. But, in spite of such discrepancies, it is possible to conclude that the releases of high-level wastes consisted of a mixture of materials from various stages of processing and with varying times of cooling. Further, it seems obvious that the radionuclide composition and physico-chemical character of the released material fluctuated widely.

In 1952–1955, an additional source of contamination became significant against the decrease of releases from the radiochemical plant. This source was reactor cooling water released into Kyzyl-Tash Lake and which subsequently flowed into the Techa River (Fig. 2). For example, during seven months in 1953, the activity released from the reactor plant was five times the release from the radiochemical plant (Fig. 8). The water of Kyzyl-Tash Lake entering the Techa was also contaminated by such activation radionuclides as ^{32}P, ^{35}S, ^{45}Ca and had a specific activity of about 7–30 kBq L^{-1} (according to measurements of 1953). For comparison, the concentration of activation products in the Columbia River downstream of the analogous Hanford plutonium-production reactors during peak operations in the early

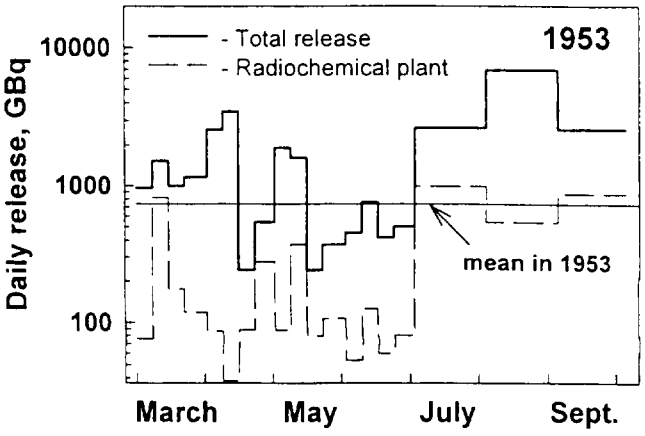

Fig. 8. Total release rates in 1953 in comparison with the release rates from the radiochemical plant.

Fn3 1960's reached an annual average[‖] of 2 kBq L^{-1} (5×10^{-8} Ci L^{-1}) and even exceeded 10 kBq L^{-1} (3×10^{-7} Ci L^{-1}) during low water flows (Wilson 1964; Napier 1993). The primary activation products at Hanford included ^{32}P, ^{51}Cr, ^{56}Mn, and ^{65}Zn. Radiation doses to downstream individuals from these radionuclides in the Columbia River averaged only a few tens of μSv y^{-1} (Napier 1993).

MEASUREMENT METHODS

The measurements of gamma-exposure rate and of radionuclide concentrations discussed in this paper are historic (early 1950's) and were obviously not made by the present authors. The methods that were used to perform the measurements have been published (Gussev et al. 1959) and will only briefly be summarized here.

Gamma-exposure rates were measured with a portable "MAK" device, which had an ionization chamber with aluminum walls as the sensing element.

The most common technique for the measurement of beta activity was the use of an end-window counter that was calibrated with an uranium-oxide standard. In order to subtract the influence of gamma rays, measurements were made with and without a 2,000 mg cm^{-2} aluminum filter that was judged sufficient to absorb all beta particles.

Total gamma activity was measured by a gas-flow counter that was placed inside a lead shield. Filters of 1.6 mm Al and 5 mm Pb were used. The count rate of samples was compared with that from a ^{60}Co source, which had been calibrated against a radium standard. As indicated above it was common to express gamma activity in terms of "gram-equivalent of radium."

Total alpha activity was measured with a flat impulse chamber with electrodes of diameter of 110 mm separated by 15 mm.

The historical methods of radiochemical separation and analysis were sufficient to permit the separation of the following five groups of radionuclides with similar chemical properties:

1. Alkaline earth elements: ^{89}Sr, ^{90}Sr, and ^{140}Ba;
2. ^{137}Cs;
3. Rare-earth elements: ^{90}Y, ^{91}Y, ^{140}La, ^{141}Ce, ^{144}Ce, and ^{144}Pr;
4. ^{103}Ru (with $^{103\,m}Rh$) and ^{106}Ru (with ^{106}Rh); and
5. ^{95}Zr and ^{95}Nb.

RESULTS AND DISCUSSION

Available environmental monitoring data

Systematic measurements of radioactive contamination in and near the Techa River started at the same time as release control (July 1951). The contamination of the river water, bottom sediments, flood-plain soils, vegetation, fish, milk, and other food stuffs, and external gamma-exposure rates were measured. Several settlements and specific sites in the upper reaches were selected as points for routine sampling. These points with historical numeration are shown in Fig. 1 (numbers 21–48) and in Fig. 9 (numbers 1–20). F9

In the 1990's, historical data of importance for dose-reconstruction purposes were arranged in the special computer database ENVIRONMENT (Degteva et al. 1996b). This database now includes more than 10,000 records of measurements of radioactive materials in the environment for the period 1951–1990: Specific alpha, beta, and gamma activity; concentrations of ^{90}Sr and ^{137}Cs in river water, concentrations of ^{90}Sr and ^{137}Cs in bottom sediments and flood-plain soils; and external gamma-exposure-rate measurements. The most representative data set was on specific beta activity of the Techa River water. Fig. 10 presents the time patterns of specific beta activity of river water at four sites in the upper reaches of the river. Significant fluctuations in concentration observed near the site of release became smoothed after the water passed through Koksharov and Metlinsky Ponds. These ponds played a very important role: They served as settling reservoirs and dampers for release peaks. As seen from Fig. 10, radionuclide concentration in water decreased one order of magnitude from 1952 to 1956. F10

The measurements of gamma- and alpha-emitter concentrations in the river water were limited in comparison with measurements of beta activity. The concentration of alpha emitters in the water of the ponds in 1952–1954 was about 4 Bq L^{-1} and decreased to 0.4 Bq L^{-1} in river water downstream of the ponds. The results of measurements of gamma activity in the river water had large fluctuations and were not correlated with beta activity (as noted above, some errors in the technique for measuring gamma activity could have existed in the early 1950's). There were also limited radiochemical analyses of the river water in 1951–1956, but these results were incomplete and sometimes discrepant. It was impossible to make any conclusions about radionuclide composition of river water on the basis of these sparse and contradictory data.

Fig. 11a, b, c illustrates the sets of data on specific beta activity of water and bottom sediments as a function of downstream distance. Such data sets were available F11a

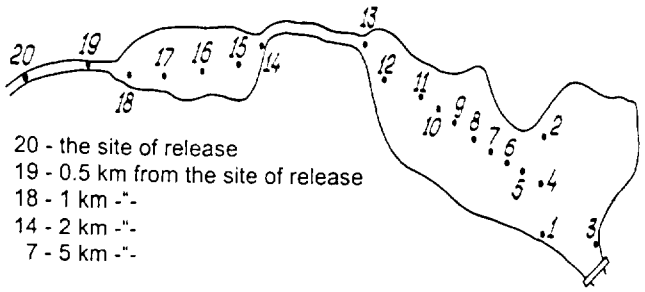

Fig. 9. Schematic map of Koksharov and Metlinsky Ponds with the sites of historical routine sampling.

[‖] For comparison, the annual average-flow rate of the Columbia River at Richland during 1950–1971 was 3400 m^3 s^{-1}.

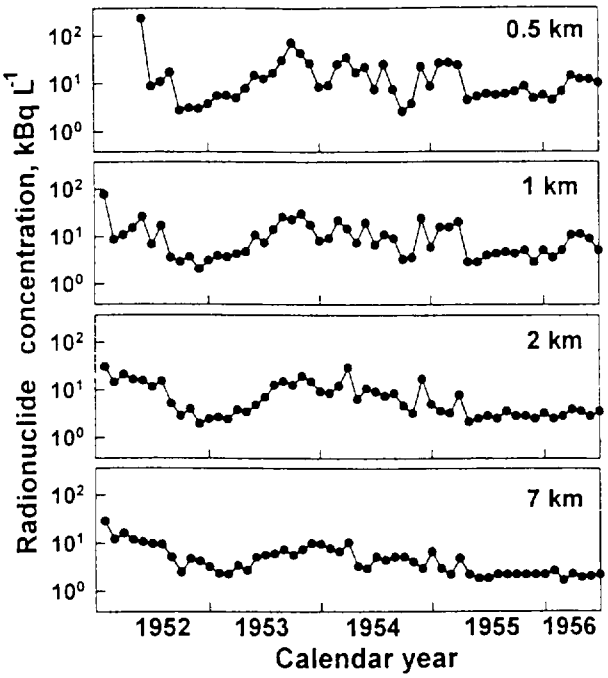

Fig. 10. Time pattern of beta activity of the Techa River water measured in the early period of contamination at several sites of the upper reaches (the range of downstream distance is from 0.5 to 7 km from the site of release).

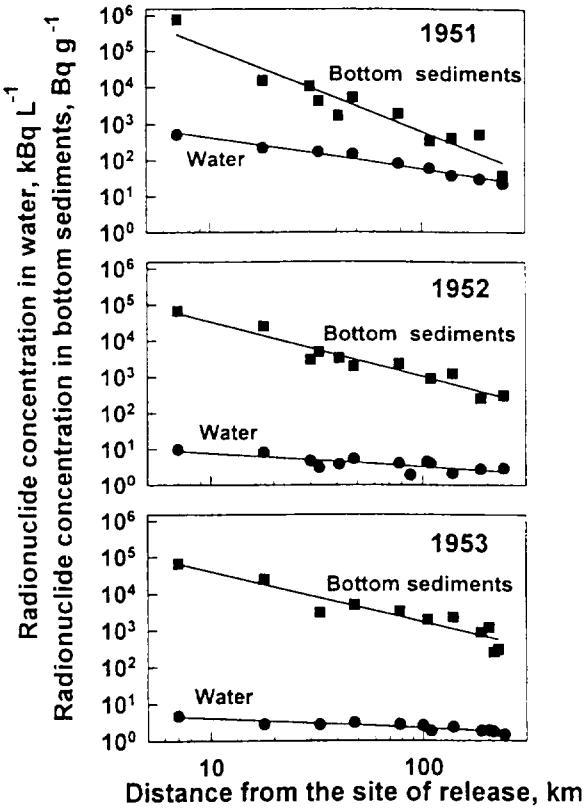

Fig. 11. Total beta activity of the river water and the bottom sediments as a function of downstream distance measured in 1951, 1952, and 1953.

for 1951–1955, and in all cases the slope of concentration vs. distance for the water was significantly lower than the slope for the bottom sediments. This means that the waterlogged bed deposit in the upper reaches of the Techa actively accumulated some of the radionuclides from the releases. Also, it is clearly apparent that the difference between contamination levels of water and sediments increases with time within the period 1951–1953: The sharp decrease of water contamination was not accompanied by a similar decrease of the contamination in the sediment, which had become a repository for long-lived radionuclides.

The first measurements of external gamma-exposure rates were performed in 1951 at several specific sites on the upper reaches of the Techa River. Starting in 1952, such measurements were performed along the entire Techa River. The results of exposure-rate measurements[¶] near the shoreline as a function of downstream distance are presented in Fig. 12. The external gamma-exposure rate did not change significantly from 1952 to 1956; this indicated rather strongly that the responsible radionuclides were long-lived (presumed to be primarily ^{137}Cs). Exposure rates measured as a function of distance from the shoreline at several sites (Fig. 13) suggested that the main source of gamma radiation was the contaminated silt, with no appreciable shielding by the water layer near the bank strip. Fig. 14 shows that there is a correlation between parallel measurements of exposure rate and beta activity of bottom sediments. It is possible to use this empirical dependence for the reconstruction of exposure rates near the shoreline for cases of known beta activity of the bottom sediments.

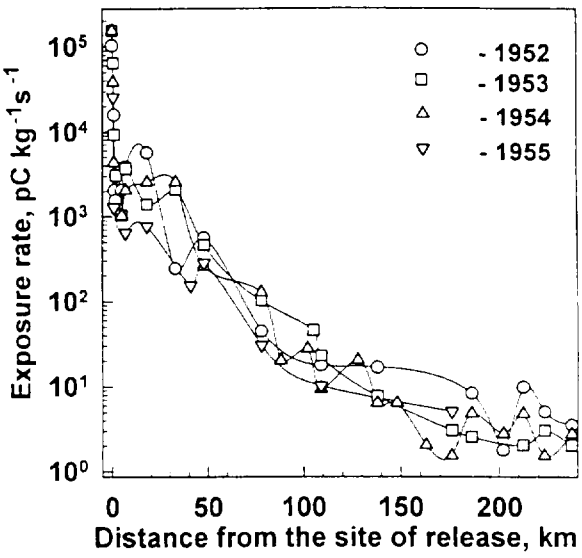

Fig. 12. Results of gamma-exposure-rate measurements near the shoreline as a function of downstream distance, as measured in 1952–1955.

[¶] The original units of measurement were $\mu R\ h^{-1}$; 1 pC $kg^{-1}\ s^{-1}$ is equal to 14 $\mu R\ h^{-1}$.

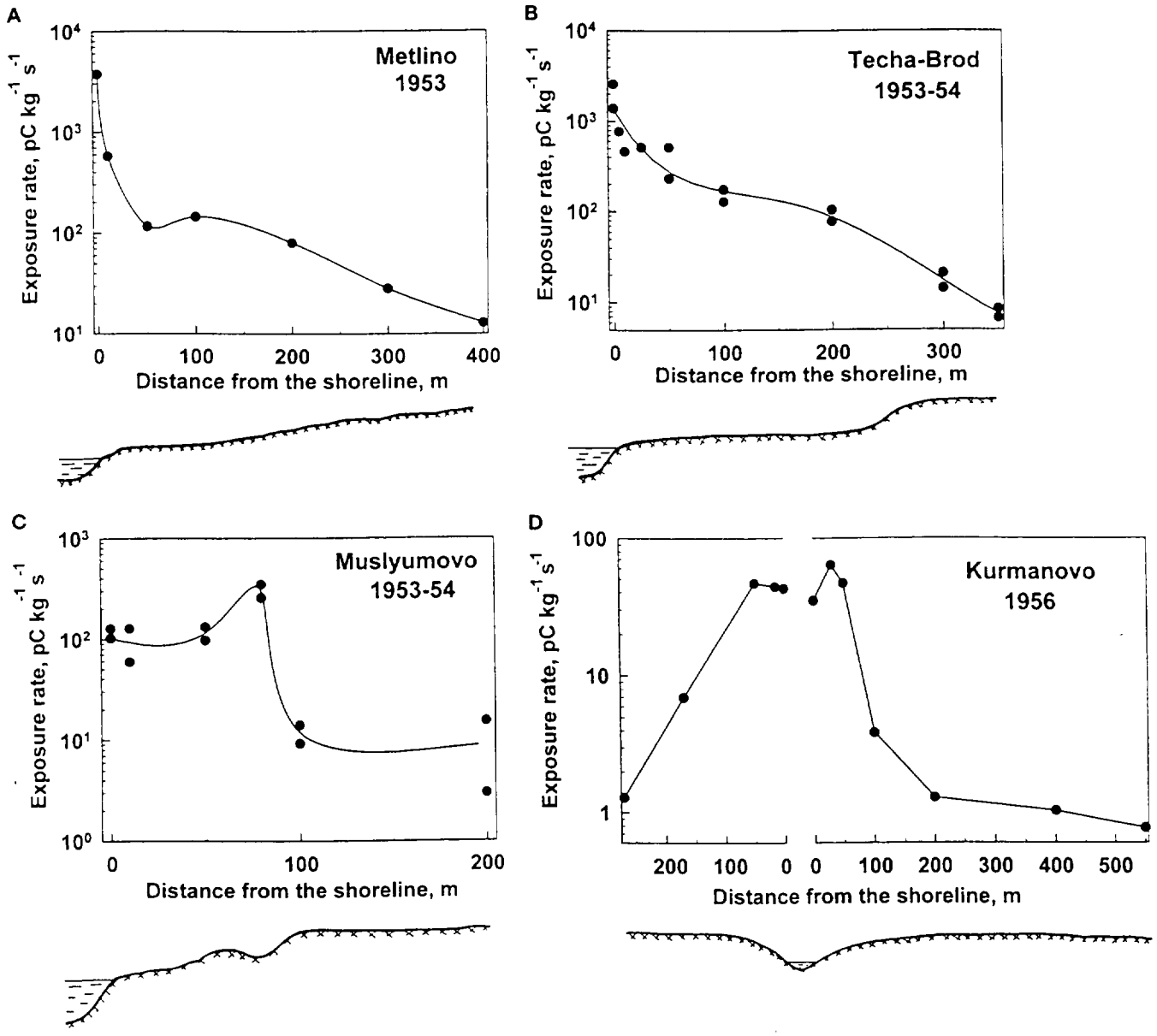

Fig. 13. Exposure rate as a function of the distance from the shoreline measured in several specific sites: (a) Metlino, 1953; (b) Techa-Brod, 1953–1954; (c) Muslyumovo, 1953–1954; and (d) Kurmanovo, 1956. In the lower part of each drawing the shore topography is indicated.

Assessment of available data

A brief description of the exposure situation on the Techa River and the data available on the source term and environmental contamination could be completed with the following considerations. Several specific periods should be considered in the evolution of the exposure situation on the Techa River (Table 4).

T4

These periods are determined by the character and completeness of data available and by the changes in the configuration of the river system. Important criteria for the determination of such stages are the magnitude of release rates and their significance for later radioactive contamination of the river system and population exposure. It is most important for the purposes of dose reconstruction to know radionuclide concentrations and exposure rates during the "acute period" of the exposure situation. Especially important was the first phase of the exposure situation, when the contact of people with the contaminated river was unlimited. As seen from Table 4, environmental monitoring data are very limited for this period. The only technique available to solve this problem is development of a model describing radionuclide transport from the site of release along the river and the accumulation of radionuclides by bottom sediments. Only a small amount of the total dose occurred after 1956, because some countermeasures, including the re-

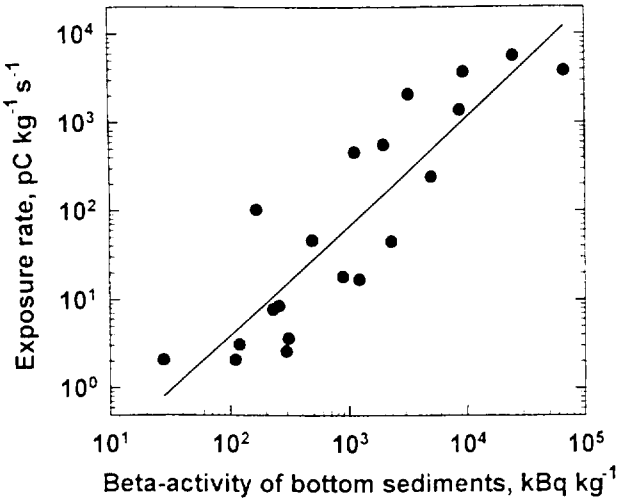

Fig. 14. Correlation of external gamma-exposure rate near the shoreline with beta-activity of bottom sediments (measurements of 1952–1953).

location of people from the upper Techa region, the ban of the use of river water for drinking and other household needs, etc., were introduced. Nevertheless, the results of later measurements of radionuclide concentrations in bottom sediments and flood-plain soils could help to validate such a river model because the contamination of the river system was caused mostly by the massive releases of 1950–1951. If the consistency between the release rates reconstructed by the Mayak experts and the environmental monitoring data could be confirmed using a more or less realistic river model, this also could serve as a test of the validity of the estimates of radionuclide releases.

In order to evaluate the presence of short-lived radionuclides and to assess the reliability of data on radionuclide composition of the releases, tables (Gussev et al. 1974) of beta- and gamma-emitting radionuclides for fission products of ^{235}U irradiated by thermal neutrons were used. First of all, the ratio of long-lived ^{137}Cs:^{90}Sr (1.05) in the radionuclide composition given by Ilyin (1956) for the massive releases of 1950–1951 is consistent with the theoretical value. This result could be interpreted as the lack of any separation between cesium and strontium in the course of chemical interactions that occurred during the process-waste transfer through a series of tanks and to the Techa River. The ratio of ^{89}Sr:^{90}Sr is 0.76, which corresponds to an age of fission products of about 1 y (with an estimated irradiation time of 120 d). The ratio of the "ruthenium group":^{90}Sr also corresponds to the theoretical value for 1-y-old waste; but this is not true for similar ratios for the "zirconium niobium" and "rare-earth element" groups. This could mean that the releases were not the "natural composition" of fission products due to chemical interactions that took place in the intermediate storage tanks. Also, it is possible and perhaps likely that the actual releases represented a mixture of several portions of fission products of differing ages. Therefore, theoretical radionuclide ratios can be used to reconstruct the presence of short-lived radionuclides only with caution and only within the groups of elements with similar chemical properties.

Similar analysis of the experimental data on the radionuclide composition for the sample of 25 September 1951 yields a value of 2.7% for ^{90}Sr and 17.5% for ^{89}Sr (on the basis of the ^{90}Sr:^{137}Cs ratio) and a value for the age of the release of about 200 d (on the basis of the ^{89}Sr:^{90}Sr ratio).

To assess the reliability of the measurements of gamma activity, the theoretical ratios of gamma-equivalent activity to beta activity (M Q^{-1}) for all fission products (given in Gussev et al. 1974) were used. It was found that the M Q^{-1} of 0.24 given by Ilyin (1956) for the massive release of 1950–1951 was reasonable: It is very close to the theoretical value for unseparated fission products with age of 150 d. The measurements for the releases of 25 September–7 October 1951 gave M Q^{-1} of 0.48, which is more than the maximal theoretical value that is obtained for unseparated fission products with a holdup time equal to 30 d. The calculations for the "experimental composition" and age equal to 150 d gave M Q^{-1} equal to 0.15, which is three times less than the reported measurement. Such a discrepancy suggests the existence of systematic errors that resulted in the overestimation of gamma activity at the time. Therefore, all early measurements of gamma activity have been excluded from the present analysis.

CONCLUSIONS AND SUMMARY

The relevant operating history of the MPA is presented. Historical data are described regarding available measurements of releases of radionuclides to the Techa River and concentrations of radionuclides in the Techa River water and sediments. The history of dam construction and watercourse changes is also described, as these alterations have had a significant impact on the movement of radionuclides within the Techa River system.

The releases during the early years (1949–1951) of operation of the MPA were the most significant and account for more than 95% of the total releases. Analysis of the available historical monitoring data indicates that the following reliable data sets can be used for reconstruction of doses received during the early periods of operation of the MPA:

- Temporal pattern of specific beta activity of river water for several sites in the upper Techa region since July 1951;
- Average annual values of specific beta activity of river water and bottom sediments as a function of downstream distance for the whole river since 1951;
- Gamma-exposure rates near the shoreline as a function of downstream distance for the whole Techa River since 1952; and

Table 4. Main stages of the evolution of the exposure situation.

Period	Main sources of contamination	Availability of source-term data	River configuration	Availability of environmental monitoring data
March 1949–October 1951: First phase of "acute period"	High-level wastes of radiochemical plant; "wild overflows"	Releases are reconstructed theoretically by the Mayak experts	Two ponds in upper reaches	Beta activity since July 1951
November 1951–November 1956: Second phase of "acute period"	Low-level wastes of radiochemical plant; Kyzyl-Tash Lake water	Release monitoring data	Two ponds in upper reaches	Beta activity, exposure rates
December 1956–1963: Isolation of the upper reach	Berdyanish Lake water; washing off EURT area	Total beta activity is evaluated	Cascade of three reservoirs in upper reaches	Beta activity, exposure rates, ^{90}Sr and ^{137}Cs since 1963
After 1963: Natural self-cleaning regime	—	—	Cascade of four reservoirs and bypass canals in upper reaches	Beta activity, exposure rates, ^{90}Sr and ^{137}Cs plutonium since 1991

- Gamma-exposure rate as a function of distance from the shoreline for several sites in the upper and middle Techa since 1951.

The results of measurements based on experimental techniques available in the early 1950's do not permit the satisfactory determination of radionuclide composition and gamma activity of the early releases from the Mayak complex. Thus, the radionuclide compositions given by Ilyin (1956), who knew the Mayak processes at that time, appear to be reasonable and the more reliable. It also seems reasonable that the presence of short-lived radionuclides can be reconstructed from theoretical ratios, but only for radionuclides within groups of similar chemical characteristics. From analysis of the data reported by Ilyin, it appears that the average age of fission products released to the Techa River in 1950–1951 was about 1 y.

Furthermore, analysis of the historical data also demonstrates that environmental modeling must be used to fill in the gaps in monitoring data.

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APPENDIX

Chronology of the main events 1948–1967: Stages in the development of the Mayak site; human interference in the Techa River system; important changes of meteorological and hydrological parameters.

Year	Time period	Event	Notes
1948	June 19	The first industrial reactor for plutonium production (A) begins operation at full capacity of 100 MW	
1948	December 22	The beginning of radiochemical plant operations	
1949	January 20–March 26	Overhaul of shutdown Reactor A	Releases of radioactive materials into the environment
1949	March	Start of liquid radioactive releases from the radiochemical plant into the Techa River	Average daily release about 2.6 TBq (70 Ci) (low-level wastes after decontamination)
1950	January	Increase of radioactive releases into the Techa River	Average daily release was about 32 TBq (860 Ci)
1950	March	Sharp increase of radioactive releases into the Techa River	Average daily release reached about 159 TBq (4,300 Ci): • Low-level wastes without decontamination • High-level wastes after treatment and decontamination • Episodic "wild overflows" up to 3,700 TBq (100,000 Ci) d^{-1}
1950	May 15	Reactor AV-1 was put into operation	
1951	April 6	Reactor AV-2 was put into operation	
1951	July 5	First measurements of river-water activity	
1951	July	Water discharge from Kyzyl-Tash Lake	Flow-rate about 7.5 m^3 s^{-1}
1951	August 11	Dam No. 3 (Koksharov Pond) was reconstructed	Capacity of pond was increased 4-fold and reached about 400,000 m^3
1951	October 13–22	Washing of Koksharov and Metlinsky Ponds. More than 15 million m^3 of water were discharged from Kyzyl-Tash Lake	Part of radioactive contaminated sediments was moved with water to the lower part of the Techa River. Essential decrease of gamma-exposure rates was observed near Metlinsky Pond.
1951	October 28	Main technological wastes were diverted into Karachay Lake	The total radioactive releases into the Techa River decreased to about 3.7 TBq (100 Ci) d^{-1}
1951	November	Creation of external dosimetry group in the Central Research Laboratory of MPA	
1951	November 17	HW-Reactor was put into operation	
1951	December 22	Reactor AI begins operation	
1952	May	Increase of the water level in Koksharov Pond by 80–90 cm	Sharp decrease of gamma-exposure rate on pond shoreline
1952	September 15	Reactor AV-3 put into operation	
1953	May 30–June 25	Cessation of water discharge from Kyzyl-Tash Lake	Decrease of the water level and increase of gamma-exposure rates in the upper Techa region
1954	March 13–April 17	Discharges of Kyzyl-Tash Lake water into the Techa River	
1954	July	High air temperatures and drought	2–3 times decrease of river-flow rate in comparison with June 1954
1955	January–March	Discharge of Kyzyl-Tash Lake water with the purpose of exchange of water in the lake	Discharge rate to the Techa was about 2.6 TBq (70 Ci) d^{-1} and total activity about 174 TBq (4,700 Ci)
1956	October–November	Dam No. 10 was constructed	Creation of Reservoir No. 10 (Shubinsky Pond). Decrease of radionuclide ~~entrance~~ release to the river system
1957	September 29	Chemical explosion of high-level waste-storage tank (Kyshtym accident)	Formation of the East Urals radioactive trace (EURT)
1958	January 1–April 25	Berdyanish Lake (volume 7.9 million m^3 with average activity 15 kBq L^{-1}) was released into Reservoir No. 10. After that more than 1.8 million m^3 of clean water from Irtyash Lake were passed through Berdyanish Lake to Reservoir No. 10.	Total beta activity released was about 127 TBq (3,440 Ci). The surface level of Reservoir No. 10 increased by 107 cm. Beta activity of water increased from 1.5 up to 15 kBq L^{-1}.
1958	April	Strong filtration through Dam No. 10 was found	Strengthening of Dam No. 10 on April 29–30
1964		Dam No. 11 and bypass canals were constructed	Creation of Reservoir No. 11 (capacity about ~~217~~ 217 × 10^6 m^3). Isolation of more contaminated upper Techa from rest of river system
1967	April 10–May 15	Wind transfer of radioactive materials from Karachay Lake	Formation of so-called New Trace

(looks funny)

APPENDIX 2

Published Article

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Paper

ISSUES IN THE RECONSTRUCTION OF ENVIRONMENTAL DOSES ON THE BASIS OF THERMOLUMINESCENCE MEASUREMENTS IN THE TECHA RIVERSIDE

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Abstract—**The potential of thermoluminescence measurements of bricks from the contaminated area of the Techa river valley, Southern Urals, Russia, for reconstructing external exposures of affected population groups has been studied. Thermoluminescence dating of background samples was used to evaluate the age of old buildings available on the river banks. The anthropogenic gamma dose accrued in exposed samples is determined by subtracting the natural radiation background dose for the corresponding age from the accumulated dose measured by thermoluminescence. For a site in the upper Techa river region, where the levels of external exposures were extremely high, the depth-dose distribution in bricks and the dependence of accidental dose on the height of the sampling position were determined. For the same site, Monte Carlo simulations of radiation transport were performed for different source configurations corresponding to the situation before and after the construction of a reservoir on the river and evacuation of the population in 1956. A comparison of the results provides an understanding of the features of the measured depth-dose distributions and height dependencies in terms of the source configurations and shows that bricks from the higher sampling positions are likely to have accrued a larger fraction of anthropogenic dose from the time before the construction of the reservoir. The applicability of the thermoluminescent dosimetry method to environmental dose reconstruction in the middle Techa region, where the external exposure was relatively low, was also investigated.**
Health Phys. 75(6):574–583; 1998

Key words: thermoluminescent dosimetry; accident analysis; environmental assessment; dose, external

INTRODUCTION

THE TECHA river and its adjacent territories (Southern Urals, Russia) were contaminated as a result of the releases of radioactive wastes by the Mayak plutonium facility from 1949 through 1956. The residents of the Techa riverside settlements who lived in the period of massive releases were exposed to both external and internal radiation. Long term epidemiological studies of the exposed population suggest that the risks of mortality from leukemia and other cancers increase with increasing radiation dose (Kossenko et al. 1997). The Techa River Dosimetry System Project was established in the Urals Research Center for Radiation Medicine (URCRM) to perform individual dose assessments by taking into account all available data sets on human and environmental contamination, as well as new data to be obtained by electron paramagnetic resonance (EPR) and thermoluminescence (TL) methods (Degteva et al. 1996). The TL technique is planned to be used mainly for the validation of calculated doses at specific sites along the banks of the Techa river within several meters of the water. Such validation is very important because the main source of gamma radiation was the contaminated silt along the river banks (Degteva et al. 1994).

TL methods have been used earlier for assessments of external gamma doses in Hiroshima and Nagasaki (Maruyama et al. 1987; Haskell et al. 1987), in the areas of the Nevada Test site (Haskell et al. 1994), in the town of Pripyat, and in the 30 km exclusion zone of the Chernobyl event (Hütt et al. 1993; Stoneham 1995). However, all these situations were significantly different from the Techa river contamination, and the study of the Ural samples could also be of importance for the further development of TL as a retrospective dosimetry method.

A pilot TL study of brick samples from the village of Metlino, located at 7 km from the site of radioactive release, has demonstrated the applicability of this technique for dose reconstruction in the upper Techa river area, where external dose rates near the river were extremely high (Bougrov et al. 1995). Further, Monte Carlo simulations of radiation transport can be used to relate source distributions of gamma radiation to depth-dose profiles in brick walls (Meckbach et al. 1996), as well as to the doses accrued in bricks at different sampling heights. For a brick sample from Metlino, the comparison of a depth dose profile determined by TL measurements with the results of Monte Carlo simulations for an assumed source distribution (Göksu et al. 1996) showed the potential of a combination of TL

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measurements with Monte Carlo simulations for obtaining information about the past distribution of radiation sources. These studies also help to formulate the issues that should be solved in the further investigations of the Techa valley.

The first issue is the evaluation of the accumulated doses due to the natural radiation in the samples. This task is important because the majority of the houses of the inhabitants of the upper Techa were demolished after the evacuation of the population in 1956, and only several massive old brick buildings, constructed a long time before the accident, are available now. Therefore, the natural radiation dose accumulated in the bricks is relatively high and special attention must be taken to evaluate carefully this contribution to the total TL dose.

The second issue is related to the changes in radiation source configuration that occurred at the Metlino sampling site as a result of the construction of an artificial reservoir in the upper Techa (the so called reservoir No. 10). This reservoir was created in 1956 after the evacuation of the inhabitants of Metlino. In order to obtain information for the reconstruction of external doses of the population from the results of TL measurements, it is necessary to find ways to determine the component of dose accrued in bricks before 1956 due to the radionuclides released by Mayak, separating it from the component accumulated after the reservoir was built. The source distribution was different during these two periods. The dependence of accumulated doses on the height of the sampling position and depth-dose distributions can be determined by TL measurements. After subtraction of doses due to natural radiation, a comparison with Monte Carlo simulations could give indications on the contribution of different source configurations to doses accrued in the bricks.

All former preliminary TL studies have been carried out only for the Metlino village in the upper Techa region, at a short distance from the site of radioactive release. A further issue is to check the applicability of TL methods for the lower parts of the Techa where external dose rates were relatively low (Degteva et al. 1994). The population of this region has received substantial doses to bone tissues due to ingestion of ^{90}Sr with river water, but the evaluation of external doses for these people is also important in order to estimate the risk of solid tumors.

MATERIALS AND METHODS

Method of dose assessment

Some minerals show thermoluminescence after having been exposed to ionizing radiation, i.e., they emit light during heating. When a recently fired material is exposed to an anthropogenic ionizing radiation field it acquires an excess dose over that which is due to natural radiation sources. The total dose accumulated in a building brick can be assessed using minerals like quartz and feldspar incorporated in it. The external gamma-dose component of the anthropogenic dose D_{Ant} can be estimated by use of the following equation:

$$D_{ant} = D_{TL} - D_{Nat}$$
$$D_{nat} = A \times (R_\alpha + R_\beta + R_\gamma + C), \quad (1)$$

where D_{TL} = total accumulated dose as measured by TL (mGy); D_{nat} = total accumulated dose due to natural radiation; A = age of the building in years; and R_α = contribution of the α-radiation of the uranium and thorium in the brick to the dose rate. This value includes a correction factor for TL efficiency of α-particles and is termed the *internal effective alpha-particle dose rate* (mGy y^{-1}); R_β = internal beta-particle dose rate due to the uranium, thorium, and potassium content of the brick (mGy y^{-1}); R_γ = dose rate due to γ−radiation of uranium, thorium, and potassium in the brick and in the environment (mGy y^{-1}); and C = dose rate due to cosmic rays (mGy y^{-1}).

Description of the sampling sites

The samples were collected at two sites at the Techa river: 1) At the upper Techa, in the former village of Metlino, located at 7 km from the site of release; and 2) at the middle Techa, in the village of Muslyumovo, at a distance of 78 km from the site of release. Bricks from three buildings located on the banks of the river were investigated: the mill of Metlino and the mill and the waterworks of Muslyumovo. For each building, two kinds of samples were collected: exposed samples taken from the outside walls facing the river and background samples taken from the inner walls of the buildings or from walls opposite to the river. The background samples were used for an assessment of the age of the sampling site. The exposed samples were used for the reconstruction of the anthropogenic dose. A description of the samples is given in Table 1.

The Metlino mill site is shown in Fig. 1, where the sampling positions on the brick wall facing the reservoir No. 10 are indicated. The samples No. 26 and No. 33 were taken from the wall at respective heights of 2 m and 4 m above the water surface and at a distance of about 4 m from the shore of the reservoir; and samples No. 32 and No. 34 were taken at a height of 6 m, at respective distances of 2 m and 4 m from the shore. Also indicated in Fig. 1 is the position of sample No. 16, investigated in

Table 1. Description of the samples.

Sample code	Location	Building	Position	Height (m)	Exposed or background
1	Muslyumovo	mill	outside wall	1	exposed
3	Muslyumovo	mill	outside wall	1	exposed
6	Muslyumovo	mill	inner wall	1	background
7	Muslyumovo	waterworks	outside wall	1.4	exposed
9	Muslyumovo	waterworks	partially destroyed wall	4.5	background
26	Metlino	mill	outside wall	2	exposed
31	Metlino	mill	inner wall	3	background
32	Metlino	mill	outside wall	6	exposed
33	Metlino	mill	outside wall	4	exposed
34	Metlino	mill	outside wall	6	exposed

Fig. 1. The wall facing reservoir No. 10 of the mill in the village of Metlino on the Techa river (7 km from the site of release). The numbers correspond to the codes of the investigated samples. Also indicated are samples No. 16, 23, 25, and 27, which have been described in Bougrov et al. (1995) and Göksu et al. (1996). The positions of the samples extracted from the wall are visible near the numbers.

a previous publication (Göksu et al. 1996), and of samples No. 23, No. 25, and No. 27, investigated in Bougrov et al. (1995). The background brick sample No. 31 was taken from the middle of a partially crushed -m-thick inner wall of the Metlino mill.

Fig. 2 shows the Metlino site in a vertical cross section (A) and a schematic view from above (B); the sampling positions on the brick wall are indicated. Before the construction of reservoir No. 10 in 1956, the Techa river passed from Metlinsky pond through a lock (partially visible at the left side of Fig. 1) and flowed at a distance of about 10 m from the wall from which samples were taken. At that time, presumably the main sources of radiation were the contaminated sediments in the river bed and its contaminated shores. After 1956, the construction of reservoir No. 10 raised the water level by about 1 m, the river flowed directly through the lock into the reservoir, and the mill stood partially in the water. Presently, the reservoir has a depth of about 1 m in the vicinity of the buildings. Close to the wall of the mill there is a narrow fringe of shallow water with a depth between 20 cm and 50 cm. Gamma dose rate measurements were performed in 1997 above the water at a distance of about 3 m from the shore at different distances from the wall, as indicated in Fig. 2b. At 1 m distance from the wall, a dose rate of about 5 μGy h^{-1} was measured; at 3 m distance the dose rate was only about 0.6 μGy h^{-1}. Farther away, the dose rates were at a level of about 0.4 μGy h^{-1}, an order of magnitude less than near the wall. On the shore of the reservoir next to the wall the contemporary dose rates range between 9 y h^{-1} close to the wall and about 3 μGy h^{-1} near the κ. The dose rate measurements indicate that the contaminated shore and the ground of the reservoir near the wall, where the water is more shallow, could have

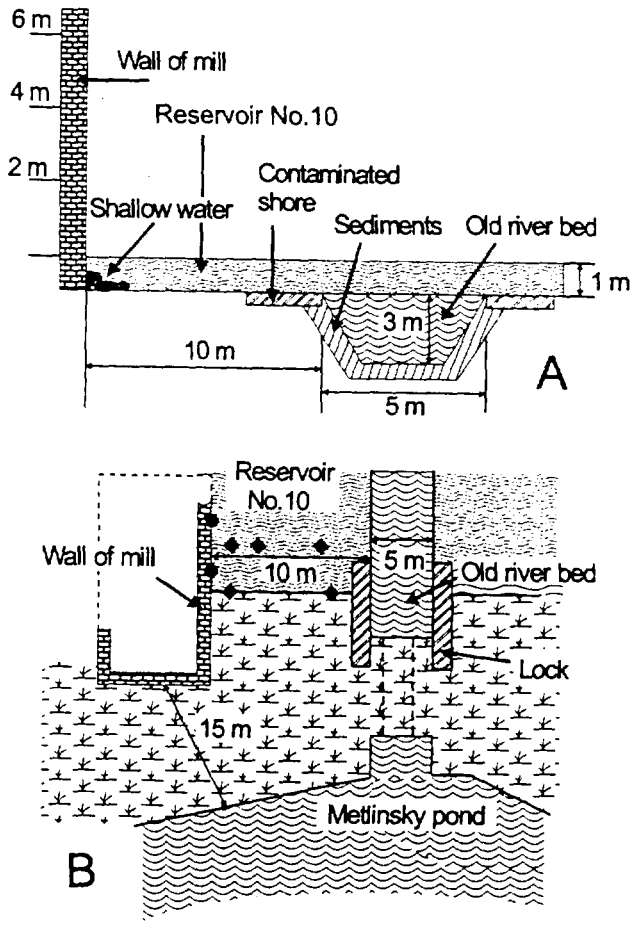

Fig. 2. Schematic view of the Metlino site, showing the position of the wall of the mill facing the reservoir (see Fig. 1) relative to the old Techa river bed: vertical cross section (A) and view from above (B). Also indicated in (B) are the sampling positions of bricks (full circles) and the locations where dose rate measurements were performed (full diamonds).

given the dominant contribution to the anthropogenic doses accrued in the bricks since 1956.

At the Muslyumovo mill, exposed samples No. 1 and No. 3 were taken from the outside wall facing the river bank (Fig. 3). Background sample No. 6 was collected at about 1 m from the outer surface of a 1.5-m-thick external wall opposite to the river. At the Muslyumovo waterworks, exposed sample No. 7 was taken from the outside wall facing the river (Fig. 4), at a height of 1 m above ground level and about 5 m above the level of the Techa river. Background sample No. 9 was extracted from the middle of the crushed round wall of the waterworks directed away from the river (Fig. 4).

Sample preparation for TL measurements

Two methods were used for sample preparation. Samples No. 9, 32, 33, and 34 were prepared using the TL quartz inclusion technique (Haskell et al. 1987) in the Center for Applied Dosimetry, University of Utah, under controlled lighting conditions. The first 3–5 mm of the

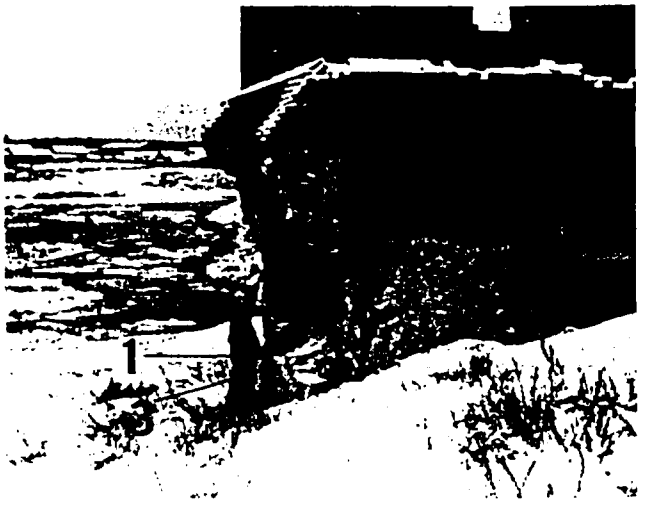

Fig. 3. The mill in the village of Muslyumovo on the Techa river (78 km from the site of release). The positions of exposed samples No. 1 and No. 3 are indicated.

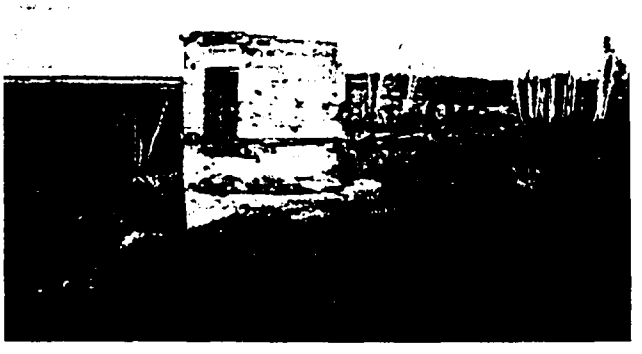

Fig. 4. The waterworks in the village of Muslyumovo on the Techa river (78 km from the site of release). The positions of exposed sample No. 7 and background sample No. 9 are indicated.

outer surfaces of each brick sample was removed using a water-cooled diamond saw, and then the samples were prepared by crushing in a hydraulic press. Particles in the size range of 106–150 μm were washed in concentrated HCl at 30°C for 1 h in an ultrasonic bath and then washed in distilled water. Then the grains were etched in 49% HF for a period of 30 min in order to remove alpha-irradiated regions of the grains, washed in distilled water, rinsed in acetone, and dried in an oven at 70°C for several hours. If precipitate was observed during microscopic examination the sample was additionally treated in HCl for 60 min. The dry crystals were separated from iron-containing particles with a magnetic separator. The non-magnetic portion of the grains was used for the measurements.

Samples No. 1, 3, 6, 7, 26, and 31 were prepared using fine-grain, additive dose or pre-dose techniques (Aitken 1985) in GSF-Institut für Strahlenschutz. The fine grain samples were prepared under laboratory red light using the Lee filter No. 106 (primary red). The outer 3 mm from all surfaces of the fragment of the brick were removed with a water-cooled diamond saw. Brick No. 26 was cut into 13 segments (1 cm thick) for the depth-dose distribution.

TL measurements

At GSF the TL glow curves were measured using an automatic reader[§] with heating rate of 5°C s^{-1} in nitrogen flow of 4 L min^{-1}. The heat-absorbing filter HA-3 was used together with Blue (Corning) BG-38 or Hoya U-340[‖] and at the low doses Corning BG-12.[¶] TL dose evaluation was made using the additive-dose method. Additive doses were given using a ^{90}Sr-^{90}Y beta ray source, which was calibrated with respect to ^{60}Co gamma ray source at the Secondary Standard Dosimetry Laboratory in GSF. The procedures of calibration are described elsewhere (Göksu et al. 1995).

The samples were stabilized at 100°C for 100 s after irradiation and before TL measurements. The short term stability of the signal was tested with the "plateau-test" (Aitken 1985) as shown in Fig. 5 for exposed sample (A) and background sample (B). The procedures of determining the accumulated dose are described elsewhere (Göksu et al. 1996).

TL measurements at the University of Utah were carried out on a Daybreak/Utah 100 TL reader[#] equipped with a 9635QA photomultiplier tube and a 40 mCi ^{90}Sr-^{90}Y beta source.** TL emission was filtered with a 4-69 filter and a 7-59 filter.[¶] TL procedures included the pre-dose technique with zero glow monitoring (Aitken et al. 1979) on Samples 32, 33, and 34.

Assessment of dose rate due to natural radionuclides

Internal effective alpha particle dose rate (R_α). The uranium and thorium content of the bricks was measured using a 4.5-cm-diameter ZnS screen with the thick-sample alpha counting method, calibrated by using the U.S. Geological Standard (BCR-1). The internal effective alpha particle dose rate was calculated using the a-value system developed by Bowman and Huntley (1984), which takes into account the efficiency of the alpha-particles independent of their energy for producing TL. The irradiation was performed with 6 plaque sources

[§] TL-DA12, RISØ Danish National Laboratory, Roskilde, DK 4000. Equipped with PM tube with bialkali photocathode (Thorn-EMI 9235QA) and optical filters. Optical filters are also available from Schott Glaswerke Hattenbergst 10, D-6500 Hainz 1, Germany.

[‖] Hoya, 7-5 Naka-Ochia 2-chome, Shinjuku-ku, Tokyo 161-8525, Japan.

[¶] Corning glasses, available from Kopp Glass Inc., 2108 Palmer Street, Pittsburgh, PA 15218.

[#] Custom-manufactured to University of Utah specifications by Daybreak Nuclear and Medical Systems, Inc., 50 Denison Drive, Guilford, CT 06437.

** Isotope Products Laboratories, 1800 North Keystone Street, Burbank, CA 91504.

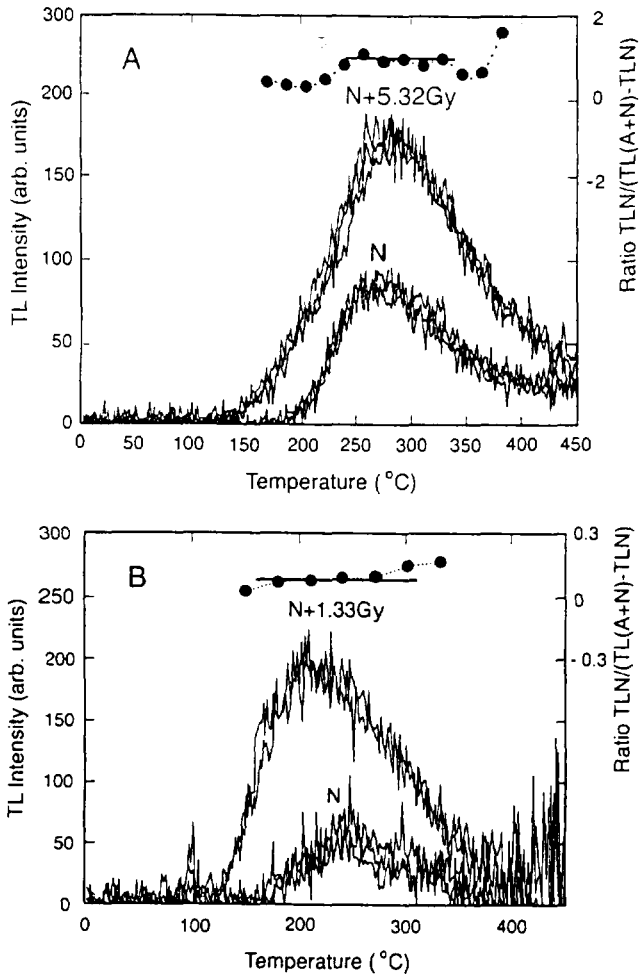

Fig. 5. TL glow curves and plateau tests for exposed (A) and background (B) samples.

with a nominal ^{241}Am activity of 6.66 GBq each and calibrated individually in vacuum.

Internal beta-particle dose rate (R_β). The Berthold LB770 β-counter[††] was used to measure the β-particle dose rate due to the uranium, thorium, and potassium content of the bricks. The thick-source beta-counting method was used for measurements and assessment (Sanderson 1988). Samples of known uranium, thorium, and potassium content were used as standards. The potassium content of the samples was also measured by gamma-spectrometry using Canberra[‡‡] bore whole detector of pure germanium (relative efficiency 55%) calibrated with $CaCO_3$ of the same geometry as a second control. Total beta-dose rates were once more calculated using uranium, thorium, and potassium contents of the samples.

[††] EG & G Schweigerweg 69, D85570 Mark Schwaben, Germany.

[‡‡] Model GCW, Canberra-Packard GmbH, Rober Bosch Str 3263303 Dreieich, Germany.

Gamma dose rate (R_γ). The internal gamma dose rate was calculated using the uranium, thorium, and potassium contents of the bricks, assuming an infinite brick media. This assumption is valid for bricks sampled from the inner parts of the 1-m-thick interior walls. For brick samples taken from outside walls, however, the internal gamma dose rate is reduced due to the air-brick interface. On the other hand, for these samples there is a contribution to the gamma dose rate from gamma radiation originating from natural radionuclides in the environment, and one expects a partial compensation of both effects. These data require further validation. Gamma ray spectrometry measurements were carried out in Utah according to the method of Lloyd (1976) with the sealed sample placed between two 20-cm (8-inch) NaI detectors.

Monte Carlo simulations

For the Metlino site, Monte Carlo simulations of photon transport from different source configurations to the brick sampling positions were performed using the code SAM-CE (Lichtenstein et al. 1979). The code allows for the simulation of complex three-dimensional geometry through a combinatorial geometry technique. By assigning materials of given atomic composition and densities to definite regions in space, the environment under consideration can be defined. Spectral photon fluences are scored in appropriate detection regions by an expected track length scoring method. Doses in brick are computed from the scored spectral photon fluences using mass energy absorption coefficients for photon interactions in bricks.

It is assumed that radiation originating from ^{137}Cs has given the main contribution to the anthropogenic doses accrued in the brick samples, and, correspondingly, the simulations were made for a source photon energy of 661.6 keV. In the simulation, the bricks were taken to have a density of 1.8 g cm^{-3}. Scoring regions were defined at the positions of the brick samples, extending into the brick wall to the same depth as the samples used for TL measurements. Also, spectral photon fluences were determined in scoring regions corresponding to the layers at different depths in the brick used for the experimental investigation of the dependence of dose on depth.

Separate Monte Carlo calculations were made for radiation originating from different sources, corresponding to the probable contamination patterns before and after the construction of reservoir No. 10 in the year 1956. For the time before 1956, it was assumed that the Techa river flowed at a distance of 10 m from the sampling wall, and that the radiation originated from its contaminated shore and from the sediments of the river bed (see Fig. 2). Accordingly, two source regions were defined: two 1-m-wide strips on both sides of the river with the radionuclides distributed homogeneously in the ground to a depth of 10 cm, and a 5-m-wide strip with radionuclides distributed to a depth of 10 cm in the river bottom sediments, below an (effective) water level of 50 cm.

For the time after the construction of reservoir No. 10, several separate source regions were defined: (1) a strip with the radionuclides distributed to a depth of 10 cm in the ground, corresponding to the contaminated shore of the reservoir next to the wall, (2) a 1-m-wide strip extending next to the wall below 30 cm of water, corresponding to the contaminated ground of the reservoir at the shallow water close to the wall (see Fig. 2), and (3) the rest of the ground of the reservoir, simulated with radionuclides distributed on the ground 60 cm below the water level.

It should be pointed out that for the conclusions to be derived in the present investigation from the results of the Monte Carlo simulations it is not necessary to make specific assumptions on the absolute or relative source strengths of the different source regions.

RESULTS AND DISCUSSION

Assessment of natural background dose rates

The content of natural radionuclides and the resulting internal alpha, beta and gamma natural background dose rates are presented in Table 2 for the samples analyzed using the fine-grain technique. For the samples prepared with the use of the quartz inclusion method, in which there is no alpha ray contribution to the measured TL dose, it is only necessary to include the internal beta and gamma dose rates, and the results are shown in Table 3. For both cases it is estimated that the internal dose rates are assessed with a 5% error. The natural background dose rates were calculated according to the Bell conversion tables (Bell 1979) and the revised data of Nambi and Aitken (1986).

For brick samples at positions at the air-brick interface there is a further uncertainty in the contribution of gamma radiation to the natural dose rate due to the difference of dose rate from photons originating from natural sources in the environment and the dose rate due to photons originating in the brick. It is estimated that this leads to a 10% error for the gamma component of the natural dose rate. The contribution of cosmic radiation to background dose rate is relatively small, and was taken to have a value of 0.28 mGy y^{-1} (Prescott and Stephan 1982). The value may be modified by the shielding effect of the buildings, and an uncertainty of 40% was assumed.

Table 2. Natural uranium, thorium, and potassium content and respective components of background dose rate for the samples prepared using fine-grain technique.

Sample No.	Uranium (ppm)	Thorium (ppm)	Potassium (by weight) %	Alpha dose rate (mGy y^{-1})	Beta dose rate (mGy y^{-1})	Gamma dose rate (mGy y^{-1})
1	1.66	4.41	1.44	0.64	1.34	0.71
3	2.48	4.61	1.52	0.84	1.52	0.83
6	2.98	6.29	1.59	1.05	1.69	0.99
7	2.04	7.57	2.00	0.92	1.87	1.03
26	2.16	4.59	1.15	0.76	1.22	0.72
31	2.38	3.50	1.22	0.74	1.27	0.86

Table 3. Internal components of background dose rate for the samples prepared with the use of quartz inclusion method.

Sample code	Beta dose rate (mGy y^{-1})	Gamma dose rate (mGy y^{-1})
9	1.31	0.80
32	1.08	0.58
33	1.89	0.96
34	1.31	0.80

TL dating of background samples

The total accumulated absorbed dose measured using TL in the so-called background bricks is assumed to be due only to natural exposure. This assumption is valid for the samples collected from the mills at Metlino and Muslyumovo, where the walls at a height of 1 m are about 1–2 m thick and the samples were taken from the interior of the walls. Therefore, the background samples were heavily shielded from external exposures. Possible internal contamination of the mills was not taken into account in this analysis. As a result, the ages of samples were determined using eqn (1) where (D_{Nat}) is measured by TL and the natural dose rates are calculated as shown in Tables 2 and 3. As can be seen in Table 4, the TL age of the Metlino Mill is found to be 132 ± 17 y with TL additive dose technique and 125 ± 14 y using TL pre-dose technique, where the alpha dose rate is ignored due to low efficiency of alpha particles producing pre-dose effect. These results are in good agreement with historical data of the buildings. The first description of the mill in Metlino was found in the book by Choupin published in 1873 (Choupin 1873), which means that the age of this mill at the time of measurement could not be less than 123 y.

For the Muslyumovo Mill a similar evaluation yielded an age of 105 ± 10 y. This is also found to be consistent with the historical data. The first mention of the mill in Muslyumovo was found in Vershova (1899), which indicates that the age is more than 97 y. For the water works tower building the quartz extraction method yielded an age of 55 ± 10 y. No written document has yet been found about the age of the building but, according to residents of this village, the waterworks in Muslyumovo was built before 1940, which indicates an age of more than 57 y.

Table 4. The age of background samples for three investigated buildings.

Sample No.	Building	Annual dose rate (mGy y^{-1})	Measured TL dose (mGy)	Calculated age (y)
6	Muslyumovo Mill	4.01 ± 0.22	420 ± 31	105 ± 10
9[a]	Water-works	2.39 ± 0.18	132 ± 20	55 ± 10
31	Metlino Mill	3.15 ± 0.19	417 ± 45	132 ± 17
31[a]	Metlino Mill	2.41 ± 0.16	313 ± 26	125 ± 14

[a] Alpha dose rates are not included due to techniques used in these measurements.

Table 5. The results of TL investigations of exposed samples.

Sample No.	Location	TL dose (mGy)	Age of building (y)	Background dose rate (mGy y^{-1})	Background dose (mGy)	Anthropog. dose (mGy)
1	Muslyumovo	380 ± 40	105 ± 10	2.97 ± 0.20	312 ± 36	68 ± 54
3	Muslyumovo	416 ± 32	105 ± 10	3.46 ± 0.22	364 ± 42	52 ± 53
7	Muslyumovo	519 ± 28	55 ± 10	4.10 ± 0.25	246 ± 44	273 ± 52
26	Metlino	4070 ± 160	129 ± 17	2.98 ± 0.20	383 ± 57	3690 ± 170
32	Metlino	2180 ± 186	129 ± 17	1.94 ± 0.16	249 ± 39	1960 ± 190
33	Metlino	2920 ± 164	129 ± 17	3.13 ± 0.21	402 ± 60	2550 ± 175
34	Metlino	3910 ± 505	129 ± 17	2.39 ± 0.18	307 ± 47	3630 ± 507

The ages obtained in the background samples are used to assess the total natural background dose in the respective exposed samples.

TL dose assessment of exposed samples

The results of TL measurements and of anthropogenic doses obtained by subtracting the dose due to natural radiation are given in Table 5. It can be seen that the anthropogenic doses for the samples from Metlino are one to almost two orders of magnitude larger than for the Muslyumovo samples. A sharp decrease of anthropogenic dose levels with distance from the site of release was initially observed with dose rate measurements made along the river in the early 1950's (Degteva et al. 1994) and also later obtained in a pilot study of the Techa riverside population by tooth electron paramagnetic resonance dosimetry (Romanyukha et al. 1996). The present results of TL measurements confirm this dependence.

For the samples collected from Muslyumovo Mill (samples 1 and 3), the accumulated dose was measured by TL with about 10% accuracy using the fine grain additive dose technique. However, for this site the anthropogenic dose could not be resolved with an uncertainty of less then 80% due to the relatively high contribution of the natural radiation to the totally accumulated dose. A smaller uncertainty could eventually be obtained using the quartz inclusion method where about ⅓ of the annual dose due to the alpha dose rate would be eliminated. These measurements need to be repeated using quartz extraction to obtain better accuracy. The difference in doses between samples from the mill (No. 1 and No. 3) and sample from the waterworks (No. 7) can be explained by different distances from the river bank strip, which is the major source of radiation (7 m and 2 m, respectively). This information demonstrates that TL methods could provide data to reconstruct the external doses to the population of the middle Techa.

Anthropogenic dose distributions at the Metlino site—comparison of measurements and simulations

The village of Metlino was the most unfortunately situated, as it was the closest settlement to the site of radioactive release, and its approximately 1,200 inhabitants received considerable levels of external dose. In order to analyze the exposure situation at the Metlino sampling site, the results of TL measurements of the present study are supplemented with data obtained in our previous studies (Bougrov et al. 1995; Göksu et al. 1996) and combined with the results of Monte Carlo simulations.

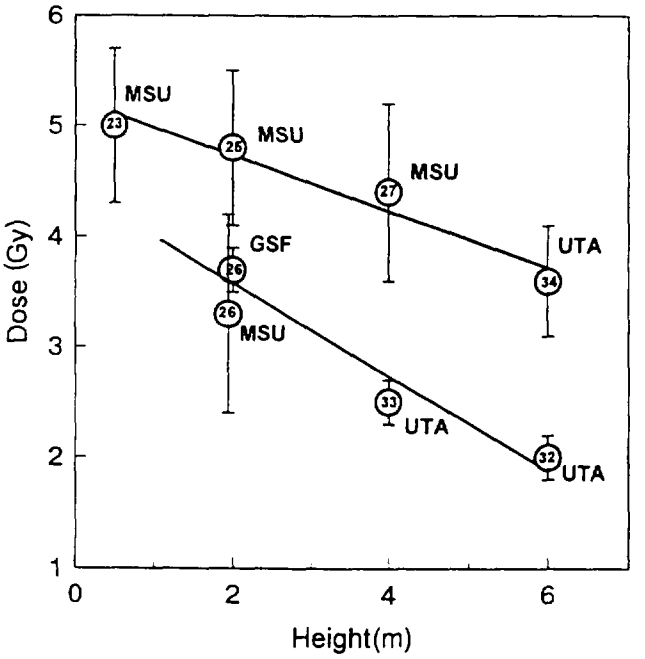

Fig. 6. Dependence of anthropogenic dose on the height of brick sampling positions for the wall of the Metlino mill (see Fig. 1). The numbers in circles correspond to the sample codes. The samples joined by the upper full line were taken at a distance of about 2 m from the shore of the reservoir; the samples joined by the lower line at a distance of about 4 m. The laboratories where the TL measurements were made are also indicated: Moscow State University (MSU); GSF-Institut für Strahlenschutz (GSF); University of Utah (UTA).

For the wall of the Metlino mill facing reservoir No. 10 (see Figs. 1 and 2), the dependence of accrued anthropogenic dose on height above the water level as obtained by TL measurements is shown in Fig. 6. Several of the samples measured in the Radiochemistry Laboratory of Moscow State University (MSU) and published in Bougrov et al. (1995) are included in the figure. For sample No. 26, an agreement within 10% was obtained for the results of measurements performed in two different laboratories. One observes in Fig. 6 that the bricks sampled at the closer distance of about 2 m from the shore of the reservoir have accrued substantially higher

anthropogenic doses than the bricks sampled at a distance of about 4 m from the shore. This could be due to a larger contribution of radiation from the contaminated shore, which is not so strongly shielded by the water. The dependence of dose on height is somewhat steeper for the samples taken at the larger distance of about 4 m from the shore; the brick sampled at a height of 2 m has an anthropogenic dose almost a factor of two higher than the brick sampled at a height of 6 m.

It is now of interest to investigate whether the dependence of accrued anthropogenic doses on the height of the sampling position can be understood in terms of what is known about the radiation source configurations before and after the building of reservoir No. 10, using the results of Monte Carlo simulations. For the samples taken at a distance of 4 m from the shore of the reservoir, Table 6 gives the ratios of anthropogenic doses measured at heights of 6 m and 4 m relative to the dose measured at a height of 2 m in comparison with results for the corresponding ratios obtained by Monte Carlo simulations for different sources. The sources considered for the time before the construction of reservoir No. 10 (river sediments and shore of the river) lead to higher doses in the upper bricks than in the lower bricks. In order to reach the lower bricks, the radiation has to effectively transverse thicker layers of sediments and water, and is therefore more strongly attenuated. The sources corresponding to the situation after the construction of the reservoir either lead to higher doses in the lower bricks than in the upper bricks (reservoir close to the wall and shore of reservoir) or to approximately equal doses (rest of the reservoir). The dependence of accrued dose on height is most pronounced for radiation from the ground of the reservoir below the shallow water close to the wall, for which the dose at a height of 6 m is a factor of five lower than at 2 m. Two factors determine this strong dependence on height: first, the strip of shallow water along the wall is narrow, so that the intensity of radiation from this source decreases nearly inversely to the height; second, at the higher sampling positions the photons effectively have to transverse thicker brick layers to irradiate the samples than at the lower sampling positions.

The results of the Monte Carlo simulations show that the source configuration before the construction of the reservoir is likely to lead to the opposite dependence of dose on height than the source configuration after the construction of the reservoir. Eventually, this feature could be used in combination with the measurement results to obtain information on doses accrued before and after the construction of the reservoir in 1956. The preliminary conclusion from comparing measured and calculated dose-height dependencies is that the anthropogenic dose in the lower bricks has a large contribution from the time after the construction of reservoir No. 10. Furthermore, the contribution to the dose from the time before the construction of the reservoir is likely to be larger in the upper bricks than in the lower ones.

The depth-dose distribution of anthropogenic dose was determined for brick sample No. 26 by TL measurements of 13 thin layers up to a depth of approximately 14 cm and subtraction of the natural radiation background dose. Results of analogous measurements for sample No. 16 have been published before (Göksu et al. 1996); for this sample, the natural radiation background dose has been reassessed, obtaining for the sample an age of 129 ± 17 y. Brick No. 26 has been sampled at a height of 2 m above the water level, sample No. 16 at a height of 1 m (see Fig. 1). The respective depth-dose distributions are shown in Fig. 7. One can see that in both cases the dependence of anthropogenic dose on depth is basically exponential beyond a depth of about 2 cm. However, the dependence is considerably steeper for sample No. 16; at a depth of 12 cm the decrease in dose is about 50% larger than for sample No. 26. The effect seems to be larger than what could be attributed to the error in the determination of the natural radiation dose background.

A qualitative understanding of this effect can be obtained by investigating the results of Monte Carlo simulations of the depth-dose distributions at the sample positions for different radiation source configurations. Also shown in Fig. 7 are depth-dose distributions calculated for sample No. 16 for contamination of the river

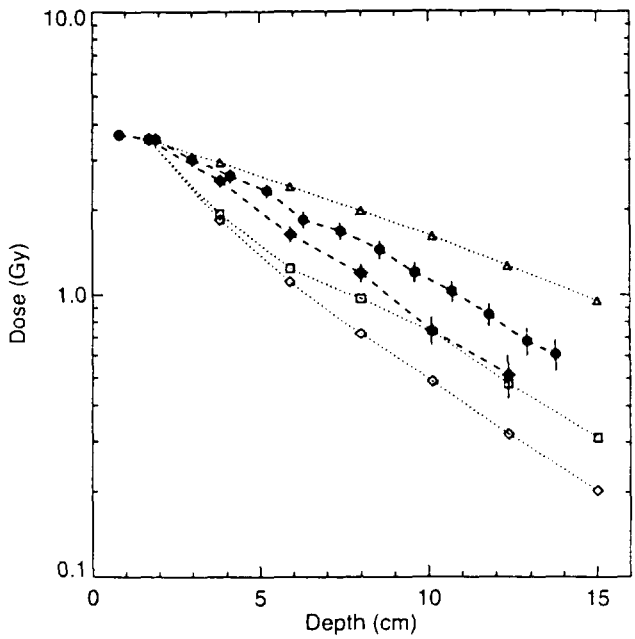

Fig. 7. Dependence of anthropogenic dose on depth as obtained by TL measurements for two bricks sampled from the wall of the Metlino Mill (see Fig. 1): Sample No. 16 (full diamonds) and sample No. 26 (full circles). Also indicated are depth dose distributions obtained by Monte Carlo simulations for radiation from the contaminated shores of the old Techa river (open triangles), from the sediments of the old river bed (open squares) and from sources in the reservoir close to the wall (open diamonds). The simulated distributions are normalized to the value measured in the first layer of sample No. 16. The dashed and dotted lines are included to guide the eye.

shores and of the river sediments at 50 cm below the water level, corresponding to the likely source configuration before the construction of reservoir No. 10, and the depth-dose distribution resulting from sources on the ground of the reservoir below the shallow water close to the wall. One can see from Fig. 7 that the reservoir source close to the wall gives rise to a very steep depth-dose profile. On the other hand, as discussed before, for radiation from this source there is a strong dependence of the accrued dose on the height of the sampling position (see Table 6). By Monte Carlo simulations, one finds that its contribution to sample No. 16, located at a height of only 1 m above the water level, is about twice as large as its contribution to sample No. 26, which is located at a height of 2 m. This could explain the steeper depth-dose profile measured for sample No. 16. Furthermore, one can see from Fig. 7 that the measured depth-dose profiles would be consistent with adequately weighted superpositions of the profiles obtained by Monte Carlo simulations for the different sources.

CONCLUSION

The results of the first steps in the Joint Russian-German-U.S. TL studies in the Techa river region are the following. Ages determined for old buildings located on the banks of the river by TL dating of background samples are in good agreement with available historical documents. Feasibility has increased for the potential use of TL methods for environmental dose reconstruction in the middle Techa region where external exposure was relatively low. For the Metlino site on the upper Techa river, anthropogenic dose accrued in bricks vs. sampling height as well as depth-dose distributions in bricks were determined by TL measurements. Several features of these distributions could be understood in terms of the past and present configurations of the radiation sources by comparisons with the results of Monte Carlo simulations. It remains to be investigated whether more refined simulations, combined with further data on depth-dose distributions and with measurements of contemporary dose rates, would allow for a quantitative determination of doses accrued at the Metlino site before the building of reservoir No. 10 and the evacuation of the population in 1956. If so, it would then be possible to reconstruct external doses in air at the river banks and use that information, which could then be used to reconstruct external doses to the population.

Table 6. Ratios of anthropogenic doses in bricks at sampling heights of 6 m and 4 m above the water level of reservoir No. 10 to the dose in brick at a height of 2 m as obtained by TL measurements in comparison with the corresponding ratios obtained by Monte Carlo simulations for different sources (see text). The source photon energy is 662 keV.

Height of sampling (m)	Ratio of doses in brick					
		Simulations				
	Measured	River sediments	Shore of river	Reservoir close to wall	Shore of reservoir	Rest of reservoir
6	0.5	1.8	1.3	0.2	0.8	1.1
4	0.7	1.4	1.2	0.4	0.9	1.0
2	1.0	1.0	1.0	1.0	1.0	1.0

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ENVIRONMENTAL THERMOLUMINESCENT DOSIMETRY MEASUREMENTS AND THEIR COMPARISON WITH VALUES CALCULATED ON THE BASIS ON HISTORICAL MONITORING DATA

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"Development of an Improved Dose Reconstruction System for the General Population Affected by the Operation of the Mayak Production Association"

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1. INTRODUCTION

The Mayak Production Association (MPA) was the first Russian site for the production and separation of plutonium. This plant began operation in 1948, and during its early days there were technological failures that resulted in the release of large amounts of radioactive waste into the rather small Techa River. The residents of the riverside communities were exposed to chronic external and internal irradiation. Extensive monitoring efforts for the environment and the population at this site began in 1951. The "Techa River Cohort" (TRC) has been studied for several decades by scientists from the Urals Research Center for Radiation Medicine (URCRM), and an increase in both leukemia and solid tumors with radiation dose has been noted (Kossenko et al. 1997). This finding suggests that, with continuing improvements in the quality of the follow-up and dosimetry, study of the TRC has the potential to provide quantitative estimates of the risks of stochastic health effects produced by chronic low-dose-rate radiation exposure in the general population. Study of this population affords an unique opportunity to address the question of the existence of a dose-rate-reduction factor for the induction of stochastic effects in an unselected general population. A definitive answer to this question would have relevance to the regulation of radiation exposure throughout the world.

The purpose of the U.S.–Russia Joint Coordinating Committee on Radiation Effects Research (JCCRER) Project 1.1 is to define and implement a protocol for improvements in the dose-reconstruction system (known as the Techa River Dosimetry System or TRDS) for the TRC, which numbers about 30,000 people (Degteva et al. 1996b). The current dose-reconstruction system is grounded firmly on whole-body counts for half of the members of the cohort (for the evaluation of internal dose, which was mainly due to incorporated ^{90}Sr) and on direct measurements of external gamma-exposure rates. This project is concerned with a comprehensive program to develop improvements in the existing dosimetry system for the TRC members by providing more in-depth analysis of existing data, further search of existing records for useful data, model development and testing, evaluation of uncertainties, verification of procedures, and validation studies of current and planned results.

A set of conceptual models that defines the relationships, pathways, and parameters that form the basis of the dose-reconstruction efforts has been described in the Final Report of a one-year pilot study (Degteva et al. 1996a). The hierarchy of information required for calculating doses to people who lived along the Techa River has also been described. One of the important tasks formulated within the framework of Project 1.1 is to perform thermoluminescent (TL) measurements for about 7–10 sites in Muslyumovo Settlement on the Techa River in order to evaluate the distribution of radiation fields. Also part of this task is the comparison of the external doses based on these measurements with the results of Monte Carlo simulations based on historical monitoring data and the results of modeling radionuclide transport in the Techa River system. It is important to stress that this task has been performed jointly with European colleagues; results are analyzed and reported for a joint pool of samples from the villages of Metlino and Muslyumovo on the Techa River.

The purposes of this document are the following:

- To present the luminescent techniques and the results of the measurements of brick samples from the Techa Riverside;
- To describe the geometry of exposure and the available historical monitoring data for each of the sampling sites;
- To present the results of Monte Carlo simulations of doses absorbed in the bricks from specific sites on the Techa River; and
- To compare the results of calculations with the results of measurements.

2. THERMOLUMINESCENCE MEASUREMENTS IN THE TECHA RIVERSIDE

Thermoluminescence (TL) methods had been used earlier to assess external gamma doses due to the explosions of atomic bombs in Hiroshima and Nagasaki (Maruyama et al. 1987; Haskell et al. 1987), fallout in the areas downwind of the Nevada Test site (Haskell et al. 1994), and fallout from the Chernobyl accident in the town of Pripyat and in the 30-km exclusion zone (Hütt et al. 1993; Stoneham 1995). However all of these situations were significantly different from that of the Techa River contamination. Thus, the study of samples from the Urals can be important also for the further development of TL as a retrospective dosimetry method.

A pilot TL study of brick samples demonstrated the applicability of this technique for the reconstruction of dose in the upper Techa area, where external dose rates near the river were high (Bougrov et al., 1995). Also, it has been demonstrated that the analysis of the dose with depth measured experimentally within a brick, compared with the results of Monte Carlo calculations, can provide valuable information about the past distribution of a specific radioactive source (Göksu et al. 1996). These studies also helped formulate the issues that should be resolved in further investigations for the Techa River Valley.

The first issue is the evaluation of the natural radiation dose accumulated by a brick; this is a function of the age of the sample. This task is important, because the majority of houses used by the inhabitants of the upper Techa were demolished after the population was evacuated in 1956. Only a few buildings (like the Metlino Mill and the Church, which were constructed long before the accident) are available now. Therefore, the natural radiation dose accumulated in the bricks is relatively high in these few surviving buildings, and special attention must be taken to evaluate carefully this contribution to the total TL dose that can now be measured in the bricks.

The second issue is the change in radiation-source configuration that occurred as a result of the construction of the artificial reservoir on the upper Techa River (so-called Reservoir No 10). This reservoir was created in 1956 after the evacuation of people. To provide information for dose reconstruction for the population living there prior to 1956, it

is necessary to reconstruct the original configuration of the radiation source and to extract the component contributed by Reservoir No. 10 after 1956.

It must noted that all preliminary TL studies were carried out only for the upper Techa region (Metlino Village, seven km from the site of radioactive release). It would be useful to check the applicability of the TL method for the lower parts of the Techa, where external dose rates were relatively low (Degteva et al. 1994). The population of this region has received substantial doses to bone tissues due to ingestion of ^{90}Sr with river water, but the evaluation of external doses for these people is also important in order to estimate properly the risk of solid tumors.

Efforts that have been taken to solve these issues are described in this report. Scientists of three laboratories from Russia, Germany and the USA are involved in the present study. Work is not yet complete; work to date is summarized.

2.1 MATERIALS AND METHODS

2.1.1. Method of dose assessment

Minerals that have been exposed to ionizing radiation will, when heated, emit light known as thermoluminescence. When a recently fired material is exposed to a transient ionizing radiation field it acquires an excess dose over and above that which can be accounted for from natural sources. The total accumulated dose received by a building brick can be assessed using minerals like quartz and feldspar, which are incorporated in the brick. In general, the external gamma-dose component of an "accidental dose," D_{Acc}, can be estimated by use of the following equation:

$$\begin{aligned} D_{Acc} &= D_{TL} - D_{Age} \\ D_{Age} &= A(R_\alpha + R_\beta + R_\gamma + C) \end{aligned} \qquad (1)$$

where: D_{TL} = Total accumulated dose as measured by TL (Gy); D_{Age} = Total accumulated dose from natural background sources due to the age of the sample (Gy); A = Age of the building in years; R_α = Internal effective alpha-particle-dose rate due to uranium and thorium content of the brick (Gy y^{-1}); R_β = Internal beta-particle-dose rate due to uranium, thorium and potassium content of the brick (Gy y^{-1}); R_γ = External gamma-dose rate from natural radionuclides at the sample position (Gy y^{-1}); and C = Dose rate due to cosmic rays (Gy y^{-1}).

2.1.2. Description of the samples

Samples were collected from two sites on the Techa River: 1) the upper Techa (former Metlino Village located seven km from the site of release); and 2) the middle Techa (Muslyumovo Village located 78 km from the site of release). The bricks from three buildings located on the banks of the river were investigated (the mill of Metlino and the mill and the waterworks of Muslyumovo). Two kinds of samples were collected for each

building: exposed samples (from the outside walls facing the river) and background samples (from the inner walls of the building or the walls opposite the river). Background samples were used to determine the age of the samples and exposed samples were used to determine the accidental dose. A description of the samples is given in Table 1.

Exposed samples No. 26 and 33 were taken from the outside wall of the Metlino Mill at 2- and 4-m height above the water surface, respectively; samples No. 32 and 34 at 6-m height from the same wall (Fig. 1). Exposed samples No 1, 3, 13 and 14 were taken from the outside wall of the Muslyumovo Mill facing the river bank (Fig. 2); and samples No. 7 and 12 were taken from the outside wall of the Waterworks facing the river (Fig. 3).

Background sample No. 31 was taken from the partially crushed thick inner wall of the Metlino Mill; this brick was extracted after removing half a meter of bricks from the crushed surface. Brick No. 6 from Muslyumovo Mill was collected from the external wall (1.5-m thick) opposite the river and about 1 m from the outer surface. Sample No. 9 was extracted from the middle part of the crushed round wall of the Waterworks and was about 90 cm above ground (Fig. 3).

2.1.3. Sample preparation for TL measurements

Two methods were used for sample preparation. Samples No. 9, 12, 13, 14, 32, 33 and 34 were prepared using the so-called TL quartz inclusion technique (Zimmerman 1971) in the Center for Applied Dosimetry, University of Utah, USA. Samples No. 1, 3, 6, and 7 were prepared using fine-grain, additive dose or pre-dose techniques (Aitken 1985) in Institut fur Strahlenschutz, GSF, Neuherberg, Germany. The fine grain samples were prepared under red light using Lee filter No. 106 (primary red). The outer 3 mm from all surfaces of the fragment of the brick were removed with a water- cooled

Table 1. Description of the samples.

Sample code	Location	Building	Position	Height (m)	Exposed or background
1	Muslyumovo	Mill	Outside wall	1	Exposed
3	Muslyumovo	Mill	Outside wall	1	Exposed
6	Muslyumovo	Mill	Inner wall	1	Background
7	Muslyumovo	Waterworks	Outside wall	1.4	Exposed
9	Muslyumovo	Waterworks	Partially destroyed wall	4.5	Background
12	Muslyumovo	Waterworks	Outside wall	5	Exposed
13	Muslyumovo	Mill	Outside wall	1.5	Exposed
14	Muslyumovo	Mill	Outside wall	2.7	Exposed
26	Metlino	Mill	Outside wall	2	Exposed
31	Metlino	Mill	Inner wall	3	Background
32	Metlino	Mill	Outside wall	6	Exposed
33	Metlino	Mill	Outside wall	4	Exposed
34	Metlino	Mill	Outside wall	6	Exposed

diamond saw. Brick No. 26 was cut into 13 segments; each segment was 1-cm thick and was used to measure the distribution of dose with depth.

The quartz-inclusion method is described in (Haskell et al. 1987). The first 3–5 mm of the outer surface of each brick were removed using a water-cooled diamond saw, and then samples were prepared by crushing in an hydraulic press. Particles in the size range of 106–150 μm were selected and washed in concentrated HCl for one hour in an ultrasonic bath and then washed in distilled water. Then, the grains were etched in 49% HF for a period of 30 minutes in order to remove alpha-irradiated regions of grains, washed in distilled water, rinsed in acetone, and dried in an oven at 70°C for several hours. The dry crystals were separated from iron-containing particles with a magnetic separator. The non-magnetic portion of the grains was used for the measurements.

2.1.4. TL measurements

The TL glow curves were measured using an automatic reader (TL-DA12, RISØ) with a heating rate of 5°C s^{-1} in nitrogen flowing at 4 L min^{-1}. A heat-absorbing filter (HA-3) was used together with a Blue (Corning) BG-38 or Hoya U-340 filter and at low doses a Corning BG-12 filter. The TL-dose evaluation was made by using the additive-dose method. Additive doses were given using a ^{90}Sr-^{90}Y beta source, which had been calibrated against the ^{60}Co-gamma source at the Secondary Dosimetry Standard Laboratory at GSF. The procedures of calibration are described elsewhere (Göksu et al. 1995).

The samples were stabilized at 100°C for 100 s after irradiation and before TL measurements. The short term stability of the signal was tested by the so-called "plateau-test" (Aitken 1985). The procedures of measurement are described elsewhere (Göksu et al. 1996).

2.1.5. Assessment of dose rate due to natural radionuclides

Internal effective alpha-particle-dose rate (R_α). The uranium and thorium content of each brick was measured using a 4.5-cm diameter ZnS screen with the thick-sample alpha-counting method, calibrated by using the U.S. Geological Standard BCR-1. The internal effective alpha-particle-dose rate was calculated using the so called a-value system developed by Bowman and Huntley (1984), which takes into account the efficiency of the alpha particles independent of their energy for producing TL. The irradiation was performed with six plaque sources with a nominal ^{241}Am activity of 6.66 GBq each and calibrated individually in a vacuum.

Internal beta-particle dose rate (R_β). A Berthold LB770 ß-counter was used to measure the ß-particle-dose rate due to the uranium, thorium and potassium content of the bricks. The thick-source beta-counting method was used for measurements and assessment (Sanderson 1988). Samples of known uranium, thorium and potassium content were used as standards. The potassium content of the samples was also measured by gamma spectrometry using a Canberra bore-hole detector of pure germanium (relative efficiency 55%) calibrated with $CaCO_3$ of the same geometry as a background control.

External dose rate (R_γ). The external gamma-dose rate is calculated using the uranium, thorium and potassium content of the bricks collected from the inner wall (1-m height) with the assumption that the most important source of gammas is within the brick.. These data require more assessment to determine the validity of the current measurements and assumptions. Bricks No. 9, 32, 33 and 34 were also analyzed by gamma-ray spectrometry. The samples were also placed between two 8-inch NaI detectors, each with three photomultipliers, and counted for 10,000 s each.

2.2. RESULTS AND DISCUSSION

2.2.1. Assessment of natural background dose rates

The content of natural radionuclides and the resulting calculated internal alpha-, beta- and gamma-dose rates are presented in Table 2. Such analyses were carried out only for the samples prepared using the fine-grain technique. It is necessary to evaluate only internal beta- and gamma-dose rates for the samples prepared with the use of the quartz-inclusion method, when the outer surface layers of grain are removed. Then, $R_\alpha = 0$. Results are shown in Table 3. The annual dose rates were calculated according to the Bell conversion tables (Bell 1979) and the revised data of Nambi and Aitken (1986). The contribution of cosmic radiation to background-dose rate is relatively small. At ground level it has been estimated to be 0.28 mGy y^{-1} (Prescott and Stephan 1982). Shielding from the building reduces this value by a factor of two.

2.2.2. TL dating of background samples

The total accumulated absorbed dose (D_{Age}) measured using TL in so-called background bricks is assumed to be due only to natural exposure. This assumption is quite valid for the samples collected from both mills at Metlino and Muslyumovo, where the walls at 1-m height are about 1–2-m thick. Therefore, the background samples were certainly heavily shielded from external exposures. As a result, the age of samples could be determined using eqn (1) where D_{Age} is measured by TL and dose rates calculated from the uranium, thorium and potassium content of the samples as shown in Table 2. As noted in Table 4, the TL determined age of the Metlino Mill is found to be 132±15 y with

Table 2. Natural uranium, thorium, and potassium content and respective components of background-dose rate for the samples prepared using the fine-grain technique.

Sample code	Uranium (ppm)	Thorium (ppm)	Potassium (%)	Alpha-dose rate (mGy y^{-1})	Beta-dose rate (mGy y^{-1})	Gamma-dose rate (mGy y^{-1})
1	1.66	4.41	1.44	0.644	1.34	0.71
3	2.48	4.61	1.52	0.835	1.52	0.83
6	2.98	6.29	1.59	1.053	1.69	0.99
7	2.04	7.57	2.00	0.920	1.87	1.03
26	2.16	4.59	1.15	0.764	1.22	0.72
31	2.38	3.50	1.22	0.742	1.27	0.86

Table 3. Internal components of background dose rate for the samples prepared by the quartz-inclusion method.

Sample Code	Beta dose rate, mGy y^{-1}	Gamma dose rate, mGy y^{-1}
9	1.31	0.80
32	1.08	0.58
33	1.89	0.96
34	1.31	0.80

Table 4. The estimated age on the basis of TL measurements of the background samples for the three buildings investigated.

Sample No.	Building	Annual dose rate (mGy y^{-1})	Measured TL dose (mGy)	Calculated age (y)
6	Muslyumovo Mill	4.01	420±31	105±8
9[a]	Waterworks	2.11	132±20	63±9
31	Metlino Mill	3.15	417±45	132±15
31[a]	Metlino Mill	2.41	313±26	125±11

[a] Alpha dose rates are not included due to techniques used in these measurements

the TL additive dose technique and 125±11 y with the TL pre-dose technique. For the latter technique the alpha-dose rate is ignored due to the low efficiency of alpha particles of producing a pre-dose effect. These results were in good agreement with historical data regarding the buildings. The first description of the mill in Metlino has been found in the book by Choupin published in 1873 (Choupin 1873). This means that the age of this mill can not be less than 123 y. The age of the Muslyumovo Mill was evaluated in a similar way with the result of 105 ± 8 y. This is also found to be consistent with historical data. The first mention of the mill in Muslyumovo has been found in Vershova (1899); this indicates that the age is more than 97 y.

The waterworks-tower building is a round tower with a diameter of 2 m and a wall thickness about 50 cm (Fig. 3). The samples inside the wall facing opposite the river were collected as background samples. The quartz-extraction method yielded a TL-determined age of 63±9 y. No written document has yet been found about the age of the building; according to residents of this village the waterworks in Muslyumovo was built before 1940; this would indicate an age of more than 57 years.

The ages obtained from the analysis of the background samples are used to assess the total background dose in the exposed samples.

2.2.3. TL dose assessment of exposed samples

The results of the TL measurements and the accidental doses obtained by subtracting the age-dose component are given in Table 5. As can be seen, the accidental doses for the Metlino samples (7 km from the site of release) are one to two orders of magnitude higher than for the Muslyumovo samples (78 km from the site of release). Such a marked decrease of exposure with distance from the source was observed initially on the basis of measurements of external gamma-exposure rates measured in the early 1950s (Degteva et al. 1994). This was confirmed later in a pilot study of the Techa Riverside population by electron paramagnetic resonance measurements of radiation dose to teeth (Romanyukha et al. 1996). The TL measurements shown here confirm this dependence.

The TL-measured accumulated dose in the samples collected from the Muslyumovo Mill (Sample No. 1 and 3) were measured with an accuracy of ±10% using the fine grain additive-dose technique. However, the level of the accidental dose is not well resolved due to the high level of age dose. A smaller level of uncertainty could be obtained using the quartz-inclusion method, where the contribution to the annual dose rate due to alpha irradiation would be eliminated. Thus, these measurements should be repeated. However, the difference in dose between the samples from the mill (No. 1 and 3) and the sample from the waterworks (No. 7) can be explained primarily by the different distances from the shoreline (7 m and 0 m, respectively). The information in Table 5 demonstrates that the TL method could provide data useful for the reconstruction of external doses to the population of the middle Techa.

Table 5. Results of TL investigations of exposed samples.

Sample No.	Location	TL dose (mGy)	Age of building (y)	Background dose rate (mGy y^{-1})	Background dose (mGy)	Accidental Dose (mGy)
1	Muslyumovo	380±40	105	2.97	312	68±40
3	Muslyumovo	416±32	105	3.47	364	52±32
7	Muslyumovo	519±28	60	4.10	246	273±28
12[a]	Muslyumovo	317	60	2.59	155	162
13[a]	Muslyumovo	395	105	2.08	229	166
14[a]	Muslyumovo	461	105	2.36	259	202
26	Metlino	4068±220	130	2.98	368	3680±220
32	Metlino	2180±186	130	1.8	234	1946±186
33	Metlino	2916±164	130	2.99	389	2527±164
34	Metlino	3910±505	130	2.26	294	3616±505

[a]The experimental errors were not evaluated for these samples, as the TL task had been cancelled before the measurements could be made.

3. AVAILABLE HISTORICAL MONITORING DATA AND MONTE CARLO SIMULATIONS OF DOSES

During the last several years, additional information on the releases and environmental contamination in the Techa River has been received from the Mayak archives (Vorobiova et al. 1999). Also, the models describing radionuclide transport in the Techa River and the gamma fields near the river shoreline have been developed (Vorobiova and Degteva 1999). The results for two specific sites representative for the upper and middle Techa region, respectively, are used in the current approach. They are the now abandoned Metlino Village located 7 km downstream of the release site and Muslyumovo Village located 78 km downstream from the release site. Metlino was the closest settlement to the site of radioactive releases at the beginning of the river contamination, and its population (1200 residents) was relocated in 1956. The population of Muslyumovo (3200 residents) has not been relocated and this settlement is now the closest non-evacuated village to the site of the past releases.

3.1. ESTIMATION OF EXTERNAL DOSE RATES NEAR SHORELINE

To evaluate external doses near the shoreline all available results of exposure-rate measurements on the shoreline for the Metlino and Muslyumovo sites were selected from the URCRM archives and data base; the results are summarized in Table 6.

Table 6. Dose rate in air near the Techa River shoreline, μGy/h.

Calendar year	Metlino site (7 km from the site of release)	Muslyumovo site (78 km from the site of release)
1949[a]	3.4	0.17
1950[a]	268–350	3.0–3.8
1951[a]	1000–1760	16.3–18.7
1952	378–567	3.2–7.9
1953	457–473	9.5–12.6
1954	252	8.5–15.8
1955	79	1.6–6.9
1958	-	2.0–6.6
1967	-	2.6
1969	-	2.2–4.4
1983	-	1.4–4.1
1984	-	2.1–3.8
1989	-	1.0–2.9
1992	-	1.1–3.0
1993	-	0.9–2.6
1998	-	0.5–2.3
Background	0.09–0.14	

[a]Only assessments with models are available for these years

Original sources of these data are described in (Vorobiova et al. 1999). Such measurements were carried out since 1952 in the summer time on the river bank near the water. For many years several different measurements have been available and in such cases the range of values is presented in Table 6. As seen, the external gamma-dose rates in Metlino (upper Techa) were two orders of magnitude higher than those in Muslyumovo Village. After the residents of Metlino Village had been relocated in 1956, the area of this settlement was partially flooded as a result of the creation of an artificial reservoir (so-called Reservoir No. 10). As seen from Table 6, dose-rate measurements at the Metlino site were stopped after 1955. Muslyumovo residents continue to live near the Techa River, but the use of river water was banned in 1953, and the flooded area was enclosed in 1956. The measurements of external gamma-dose rates continued, but the major part of the population did not receive additional external dose near the shoreline after 1956.

To reconstruct gamma-dose rates in air during 1949–1951, a model describing radionuclide transport from the site of release along the river and the accumulation of radionuclides by bottom sediments has been used (Vorobiova and Degteva 1999). This model successfully correlated the release rates of radionuclides, hydrological data, and available environmental monitoring data for the early period of contamination. Another important result of the modeling was the possibility to reconstruct external gamma-dose rates in air on the river bank in the early years, when there were no appropriate measurements (Vorobiova and Degteva 1999). Exposure rates measured in 1952–1953 as a function of distance from the shoreline at several sites suggested that the main source of gamma radiation was the radioactive silt, with no appreciable shielding by the water layer near the bank strip. The silt was contaminated predominately by ^{137}Cs, ^{95}Zr, ^{95}Nb and $^{103,106}Ru$. It is possible to use the empirical dependence of dose rate in air on beta activity of bottom sediments derived from the parallel measurements of both values carried out in 1952–1953 on the Techa River and described in Vorobiova et al. (1999). Another possibility is calculation on the basis of modeled radionuclide concentrations in bottom sediments using coefficients obtained by Monte Carlo simulations of air kerma for contaminated soil with a dose-reduction factor for river shorelines (Eckerman and Ryman 1993). The results of these calculations are described in Vorobiova and Degteva (1999) and also illustrated in Fig. 4 (a, b, c) for 1950, 1951 and 1952, respectively. For 1952 the results of measurements are also presented for comparison (Fig. 4c). Modeled dose rates in Metlino and Muslyumovo for 1949–1951 calculated by both methods are shown in Table 6. It is assumed that these values calculated using the two approaches should characterize the possible range of dose rates in 1950–1951.

The marked increase of external gamma-dose rate from 1949 through 1951 (Table 6) is determined by the increased rate of radioactive releases into the Techa River and accumulation of radionuclides by bottom sediments. After this period, variations in external dose rate are determined by natural variations of water level and artificial washings of the river by discharges of water from Kyzyl-Tash Lake, which is located upstream from the release site and separated by a dam (Vorobiova et al. 1999). In general, gamma-dose rates decreased by an order of magnitude since 1951–1953 as a result of radiation decay and natural self-cleaning processes.

3.2. CALCULATION OF ABSORBED DOSES IN BRICKS

In order to perform model calculation of dose accumulated in bricks since 1949, it is necessary to investigate some parameters specific to exposure geometry and data available for each sampling site. The general formula for calculating dose in bricks is the following:

$$D_{ant} = 8760 \cdot k_1 \cdot k_2 \cdot \sum_i P_i \qquad (2)$$

where

D_{ant} = Dose from anthropomorphic sources accrued in brick since the beginning of contamination in 1949 up to 1992 when the majority of measured brick samples were collected (Gy);
8760 = Number of hours in a year;
k_1 = Geometry factor describing the ratio of dose in the first 1-cm layer of brick-to-the dose in free air;
k_2 = Seasonal factor describing the decrease of annual dose due to shielding by snow during winter time; and
P_i = Dose rate in air near the shoreline in summer for year i (i = 1949...1992) (Gy h^{-1}).

The values of k_1 were calculated for different sampling sites using the electron-photon Monte Carlo transportcode, CASCADE-5 (Lappa and Burmistrov 1994). The value of k_2 for typical weather conditions and typical winter-snow level in the middle of Russia, including the Urals, has been evaluated as 0.85 (Balonov et al. 1995).

Specific details of the dose calculations for each sampling site are described below.

3.2.1. Metlino site

Brick samples were taken from the wall of the mill at different heights for two vertical profiles located at a distance of 2 m apart (Bougrov et al. 1998). The geometry of radiation exposure of this wall underwent a major change in 1956 after the creation of Reservoir No. 10. Therefore, it is necessary to consider separately the doses accrued before and after 1956.

Before 1956 the wall was facing the river and was parallel to the shoreline at a distance of 10 m. The main sources of irradiation were the highly contaminated shore of the river (layer of bottom sediments) and the less contaminated flooded soil between the shore and the wall. Monte Carlo simulations of height dependence of dose for two such configurations of radioactive sources lead to opposite results: higher levels in the upper bricks for irradiation from the shore and lower levels in the upper bricks for irradiation from the flooded soil. To calculate the geometry factor for the superposition of these two sources, it is necessary to evaluate the ratio between them.

According to gamma-dose-rate measurements performed in 1952–1954 near this site at different distances from the shoreline, the decrease in dose rate at a distance of 10 m and a height of 1 m is evaluated as 0.17. Monte Carlo simulations of free-in-air dose rate at 1 m above ground as a function of distance from the shoreline were performed for two source configurations. The first was for two layers of river sediments (1-m width and 5-cm depth) contaminated by ^{137}Cs and separated by the river (5-m width); these layers were covered by water with effective depth of 5 cm. The second configuration was for a zone of soil contaminated with ^{137}Cs (20-m width and 5-cm depth). The configuration of the actual source of dose in bricks was assumed to be the superposition of the "shoreline source" and the "floodplain source" that would result in the same decrease of exposure rate with distance that was measured in 1952–1954.

Monte Carlo simulations of height dependence of dose in bricks for such configurations of radioactive sources were performed, and the ratios of dose in free air at a height of 1 m-to-dose in bricks were calculated. These coefficients (Table 7) characterizing the height of the sampling position were introduced into dose calculations performed according to eqn (2) for the period 1949 through 1956.

After 1956, the construction of Reservoir No. 10 raised the water level by about 1 m, and the mill stood partially in the water. Presently, the reservoir has a depth of about 1 m in the vicinity of mill. There is a narrow fringe of shallow water with a depth between 20 and 50 cm close to the investigated wall. The main source of gamma irradiation of bricks in such a geometry is the shallow water close to the wall. Gamma-dose-rate measurements performed in 1996–1997 near each sampling position showed that the dose rate decreases with height (Table 7). The values of k_1 were calculated as

Table 7. Calculated doses in bricks of Metlino mill.

Dosimetric value	Height of sample position over the water level after 1956 (m)			
	1	2	4	6
Dose rate in air measured in 1996-1997 (μGy h^{-1})	3.0–5.0	1.9–2.8	1.0–1.3	1.3–1.4
Ratio of dose in air-to-dose in brick for exposure geometry after 1956	1.19	1.54	2.12	3.43
Calculated dose accrued in bricks after 1956 (Gy)	1.4–2.2	0.7–1.0	0.25–0.31	0.19–0.23
Ratio of dose in air-to-dose in brick for exposure geometry before 1956	1.27	1.15	0.96	0.87
Dose accrued in bricks before 1956 (Gy)	2.2–3.2	2.4–3.5	2.9–4.2	3.2–4.6
Calculated total dose accrued in bricks (Gy)	3.6–5.4	3.1–4.5	3.2–4.5	3.4–4.8

ratios between gamma dose in air near the sampling position at different heights-to-dose in bricks with the assumption that the irradiation source is ^{137}Cs in the layer of bottom sediments near the wall (effective width of 1 m, effective thickness of 10 cm) shielded by a water layer of 30-cm depth. The results of these calculations are also presented in Table 7. Finally, the comparison of summer (over the water) and winter (over the ice) measurements gives a value of 0.77 for the seasonal coefficient k_2; this is lower than typical and results from shielding by a layer of ice in addition to the shielding by snow.

Measurements of radionuclide concentration in the water of Reservoir No. 10 have been performed since 1956, but gamma-dose-rate measurements on the reservoir's shore at the Metlino site were started only in 1986 (Fig. 5). In addition, there were several measurements of radionuclide contamination of the bottom sediments. The results of these investigations have shown that the water and sediments are contaminated by ^{90}Sr and ^{137}Cs and are in dynamic equilibrium between the water and sediments. For such conditions it was assumed that the dose rates in air near Reservoir No. 10 were proportional to the ^{137}Cs concentration in water (and therefore its concentration in sediments) during the whole period of its existence (Fig. 5). Such assumptions permit the evaluation of dose accumulated during 1956–1992 using contemporary site specific measurements of dose rate and the time dependence of ^{137}Cs concentration in water:

$$D_{ant}(1956-1992) = k_1 \cdot k_2 \cdot P_{1992} \cdot \int_{1956}^{1992} e^{\lambda t} dt \ , \qquad (3)$$

where $\lambda = 0.0447\ y^{-1}$.

3.2.2. Muslyumovo site

The external dose rates at the Muslyumovo site were two orders of magnitude lower in comparison with those at Metlino, because Muslyumovo is located at a distance of 71 km farther from the site of radioactive release. The Techa River near Muslyumovo has a sloping shore and the river bed has several branches, some of which dry up during dry summers. The shoreline is located at a distance of 7 m in high water periods from the wall where samples were taken and at a distance of 19 m during dry summers. According to gamma-dose-rate measurements performed in 1952–1954 near this location and at different distances from the shoreline, the decrease in dose rate is evaluated as approximately 0.33. Late measurements (1993 and 1998) confirmed this value despite the changes in the distance from the shoreline. Monte Carlo simulations were performed for the Muslyumovo Mill in the same manner as for the Metlino Mill. Calculations of the total dose accrued in bricks of the Muslyumovo Mill using dose rates from Table 6 (with interpolation for time gaps in measurements) give a value of 0.14–0.35 Gy.

4. COMPARISON OF CALCULATED AND MEASURED VALUES

As described above, the bricks from three buildings located on the banks of the Techa River were investigated: the Metlino Mill and the Muslyumovo Mill and

Waterworks. Anthropogenic doses accrued in exposed samples taken from the outside walls facing the river and located several meters from the shoreline were determined by TL measurements (Bougrov et al. 1998). Metlino was in the most unfavorable exposure situation, because it was the closest settlement to the site of radioactive release, and the 1200 inhabitants received substantial levels of external dose. In order to analyze the exposure situation in Metlino, the results of the present study were supplemented with data presented in our previous studies (Bougrov et al. 1995; Göksu et al. 1996).

Table 8 illustrates the measured and calculated values of absorbed dose for the brick samples located at different heights on the wall of the Metlino Mill. The comparison between the measured and calculated values confirms that the dose rates in air near the shoreline as reconstructed with the river model for the first few years of contamination are reasonable; thus, the river model can be used to assess the external dose of the population.

The results of TL investigations for only two exposed bricks taken at a height of 1 m from the wall of the Muslyumovo Mill were published in our earlier paper on the feasibility of using this method for the middle Techa region (Bougrov et al. 1998). The preliminary evaluation method gave a value of 0.05–0.07 Gy for the anthropogenic doses accrued in bricks with an uncertainty level of 80–100%. This result is significantly lower than the value of 0.14–0.35 Gy obtained by Monte Carlo simulation. This discrepancy was addressed by continuing the TL investigations in Muslyumovo and by measuring samples collected at a height of more than 1 m; this study was suggested by the comments of some elderly residents of Muslyumovo that there was a wood fence between the wall investigated in our study and the river in the early 1950s. Such a fence could shield the lower part of the mill wall from "river irradiation." As seen in Table 5, anthropogenic doses accrued in bricks No. 13 and 14 located at heights of 1.5 m and 2.7 m are equal to 0.14-0.19 Gy, which is in agreement with the calculated values.

Brick samples from the Muslyumovo Waterworks were deduced to have received anthropogenic doses equal to 0.16–0.27 Gy. Unfortunately, there were no dose-rate measurements near this building during the early period of contamination. The geometry of exposure for the waterworks is complicated and different from the geometry of the mill. Nevertheless, current dose rates near the walls of investigation are almost the same for both buildings. This suggests that the time patterns of environmental exposure were

Table 8. Calculated and measured doses in bricks of the Metlino Mill.

Dosimetric value	Height of sample position over the water level after 1956 (m)			
	1	2	4	6
Calculated total dose accrued in bricks (Gy)	3.6–5.4	3.1–4.5	3.2–4.5	3.4–4.8
Measured anthropogenic dose accrued in bricks (Gy)	4.0–5.0	3.6–4.8	2.8–4.2	2.0–3.6

approximately the same in all places on the shoreline near Muslyumovo, and it could be assumed that approximately the same values of dose accrued for the bricks at both the mill and the waterworks.. On the basis of such suggestions, the TL doses measured for the bricks from the waterworks also agree with the results of calculations. However, this conclusion can only be considered as preliminary, and further investigations are needed to support the conclusion.

5. SUMMARY AND CONCLUSION

The results of the first steps in the joint Russian–German–U.S. TL studies in the Techa River region are the following.

- The ages of the old buildings located on the banks of the Techa River have been determined by "TL dating" of background samples. The results are in good agreement with available historical documents.
- The applicability has been demonstrated of the TL method for environmental dose reconstruction in the middle Techa region, where external exposure was relatively low.
- Detailed investigation of the source configuration and Monte Carlo simulations of accrued dose for each brick sample are very important for interpretation and understanding of the TL results.
- The feasibility has been demonstrated of the use of contemporary experimental methods of retrospective dosimetry for the validation of external dose calculations in the Techa River Valley

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Fig. 1. The wall facing Reservoir No. 10 of the mill in the village of Metlino on the Techa River (7 km from the site of release). The numbers correspond to the codes of the samples investigated for this report.

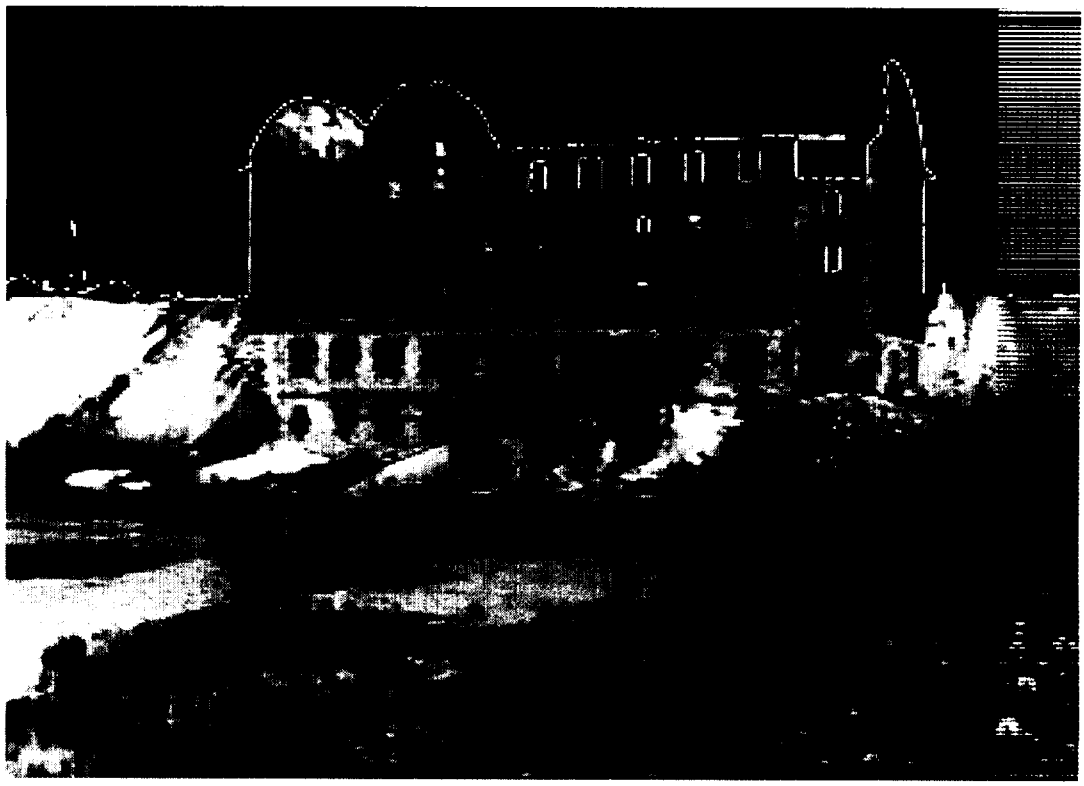

Fig. 2. The mill in the village of Muslyumovo on the Techa River (78 km from the site of release).

Fig. 3. The waterworks in the village of Muslyumovo on the Techa River (78 km from the site of release). The positions of exposed sample No. 7 and background sample No. 9 are indicated.

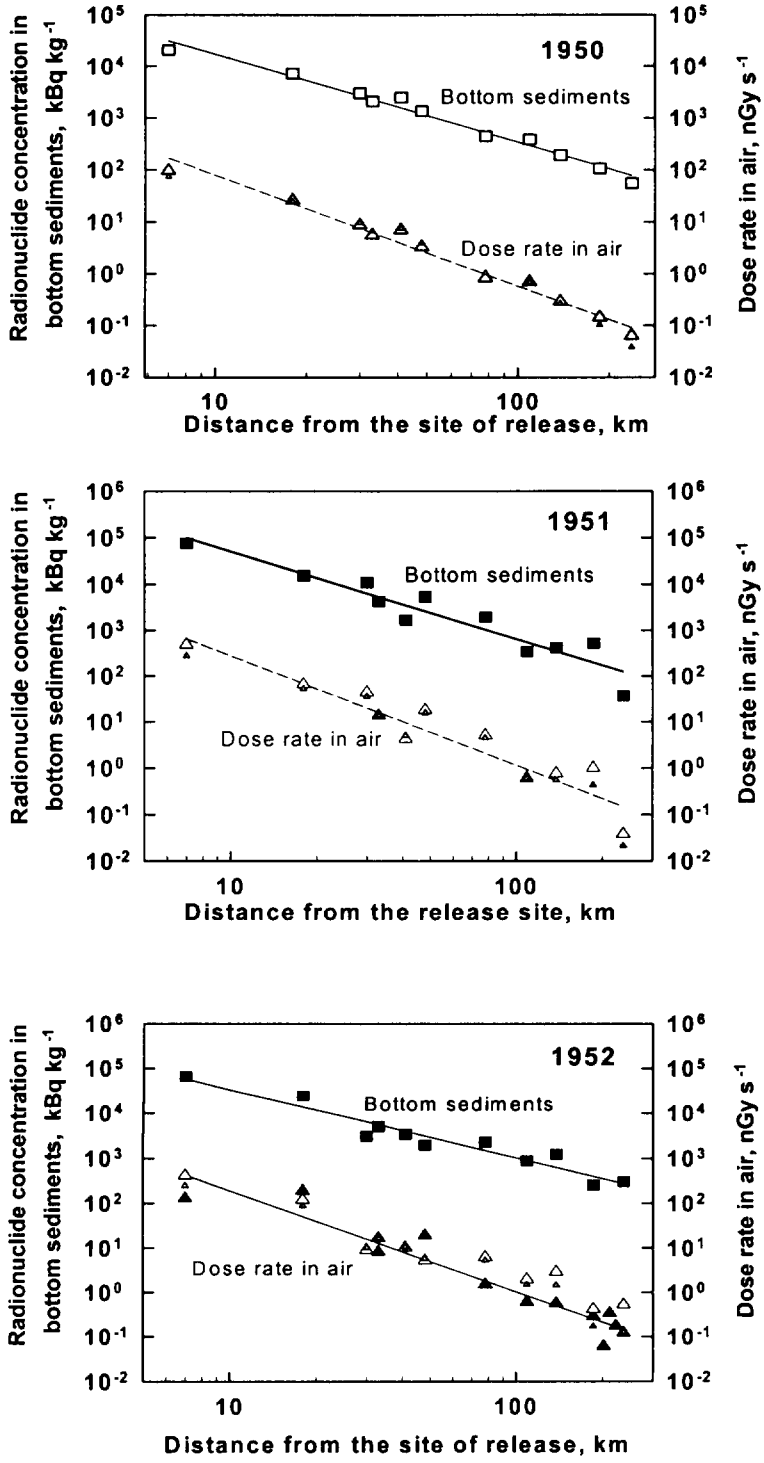

Fig. 4. Inferred external gamma-dose rate (triangles) in a) 1950, b) 1951, and c) 1952, as a function of downstream distance (black = measurements; gray = Monte-Carlo simulations; open = calculation from empirical curve). Squares correspond to beta-activity of bottom sediments (black = measurements; open = river-model calculations).

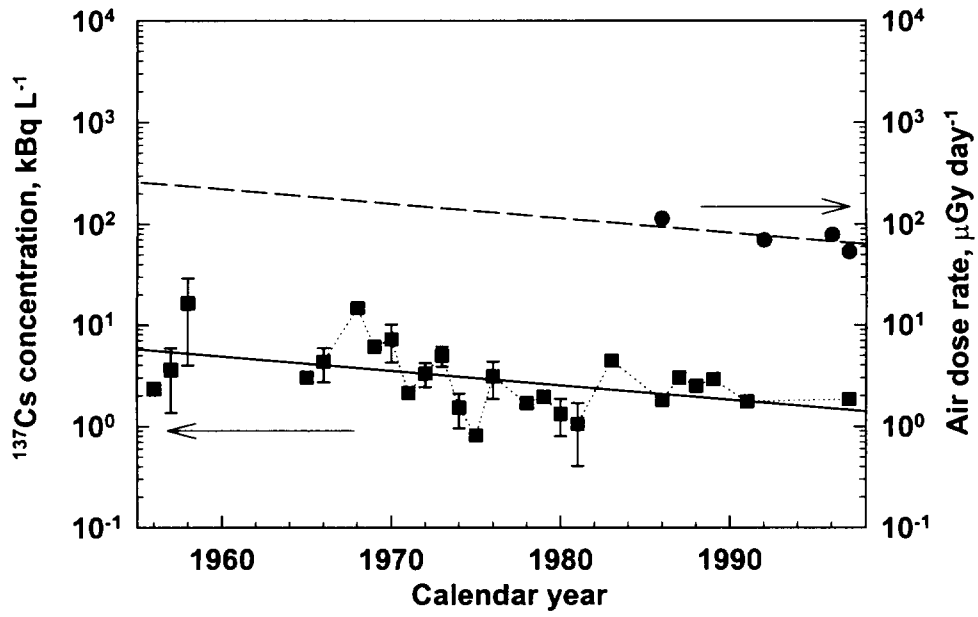

Fig. 5. Measurements of ^{137}Cs concentration in the water of Reservoir No. 10 (squares) and gamma-dose-rate measurements (circles) on the shore at Metlino.